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COMPREHENSIVE STUDY OF MARITIME AEROSOLS

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State University of New York
Scotia, New York

5 August 1971

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COMPREHENSIVE STUDY OF MARITIME AEROSOLS

FINAL REPORT CONTRACT CPA 70-64

5 AUGUST 1971

SUBMITTED TO ENVIRONMENTAL PROTECTION AGENCY

DIVISION OF METEOROLOGY

by

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ATMOSPHERIC SCIENCES RESEARCH CENTER

and

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59

Aerosol observations have been obtained along seven (7) trade routes. These observations have been performed by watch officers of merchant ships with the cooperation of the several shipping lines. Additionally, six (6) Atlantic crossings have been made during the contract year by the Training Ship Empire State IV, via a Bermuda Azores route.

Aerosol concentrations observed four (4) times a day, coincident with the NOAA synoptic observation were communicated to the investigators and cadet analysts at ASRC and the Maritime College for analysis and plotting. North Atlantic crossings require about 30 days for a complete trip, South Atlantic routes to Africa 90 days, and Indian, Australian, and world circumnavigation routes require 90 to 180 days for return of ship and data to home port. As our cooperating ships are based in New York Harbor, the maximum frequency of observations occurs in the North Atlantic. Sufficient data is already available to determine average concentrations in this region; additional crossings are necessary to obtain significant averages over the Pacific and Indian Oceans.

Lead aerosols have been detected, using the cold box technique, aboard the T. S. Empire State and in the laboratory at the Maritime College. The concentration of lead aerosol decreased rapidly with distance from the continent in July and August of 1970; the experiment is being repeated in the same area during July and August 1971.

The principal investigator was able to perform several observations in an area of the North Atlantic not on the main trade routes, aboard the Research Vessel Mètéor. While participation in the Mètéor expedition was not funded by this contract, some equipment and supplies were diverted to this operation, and a summary report of Mètéor data is included as an appendix. Also, as supplies, equipment, and travel funded by this contract helped support joint development of two new instruments for counting and sizing maritime aerosols, descriptions of these new instruments are included in appendices A and B.

TABLE OF CONTENTS

Section I. Introduction.

Section II. Area Coverage of Observer Network.

Section III. Instrumentation.

Section IV. Quality and Reliability of Data.

Section V. Recording, Transmittal and Storage of Data.

Section VI. Preliminary Data
Ice Nucleus Observations
Aitken Nucleus Concentrations at Sea
Observations in the Aleutian Islands
Continental Experiments to Support Program

Section VII. Conclusions and Recommendations.

Section VIII. Acknowledgements.

Section IX. Bibliography.

Appendices

Appendix A. A Portable Photo-Recording Aitken Nucleus Counter.

Appendix B. A Field Device for Aerosol Size Measurement.

Appendix C. Recent Aerosol Observations Over the North Atlantic.

Appendix D. Instructions for Cooperating Observers.

Appendix E. Data Used to Construct Figure 3, Section VI.

I INTRODUCTION

The objective of this research program is to obtain the distribution of aerosol concentrations over the seas. This distribution will then be used to estimate the relative strengths of several continental areas as aerosol sources, and the strength of several meteorological regimes as aerosol sinks.

Shiratori (1934) compiled a map showing the distribution of aerosol concentrations in the maritime regime, using the data obtained during the many voyages of the "Carnegie". Landsberg (1938) also analyzed the Carnegie data, and that of several other observers, and prepared frequency distributions and averages for maritime aerosol concentrations which are still frequently quoted. More recently Hess (1948, 1951), Gunn (1964) and Hogan et al, (1967) have made occasional measurements over the North Atlantic. Gunn implied the existence of a secular increase in world wide fine particle pollution from comparison of his measurements with historical electric field data.

It is desirable to establish a baseline for world wide aerosol concentrations representative of the current decade, as the Carnegie established for the first quarter of this century. We are attempting to do this by supplying aerosol instruments to watch officers on cooperating merchant ships, to allow them to perform aerosol observations concurrent with synoptic weather observations. The frequent crossings, via repetitive routes, of large, remote, oceanic areas by these ships, appears to be a rapid way to determine the aerosol distribution at relatively low cost. The frequency of these crossings will also allow the estimation of the short period fluctuation in aerosol concentration with meteorological phenomena.

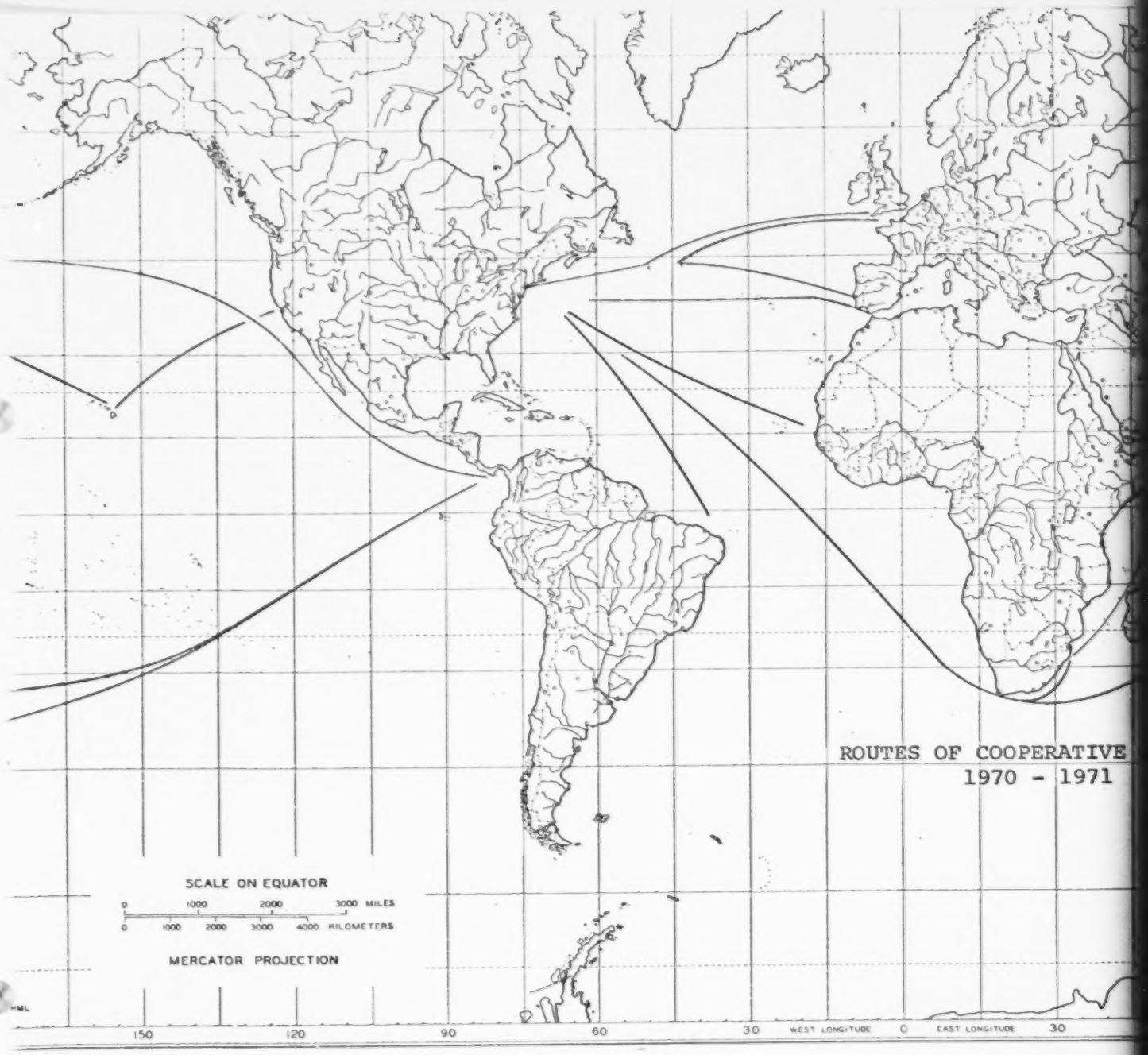
II AREA COVERAGE OF OBSERVATION NETWORK

Our initial proposal anticipated that we would be able to provide three cooperating observers, in addition to the Maritime College cadet observers aboard the Empire State IV, during 1970-71. The fine cooperation of the several shipping companies, their port personnel, and especially the deck officers, has allowed us to establish observations on seven (7) routes, in addition to the Bermuda - Azores High route of the Maritime College training cruises. The general great circle routes followed by these ships are shown in Figure I; the actual courses followed over a period of several crossings vary slightly, due to local weather and ice. A description of these routes follows.

1) New York-Northern Europe. This is the traditional North Atlantic Great Circle Trade Route, from New York to the vicinity of Cape Race, Newfoundland, often going as far as 60°N when no ice is reported and the first port of call is in Scandinavia. When ice is present, more southerly routes are taken. (North Atlantic tracks A, B, C and D, - see for example Bowditch, HO Pub. 9, pg. 758). Currently, this route is serviced by fast container ships which return to New York at about 30 day intervals. The Moore Mac Sun (Moore McCormick Lines), and the American Argosy (U. S. Lines) cooperated in this program during 1970-71. Both of these ships have recently been diverted from North Atlantic service, requiring the transfer of instrumentation. Data obtained from this route is quite interesting, as it allows examination of the penetration distance of continental aerosol in an area of frequent cloud and storm, as opposed to the cloudless area often studied further south in the Bermuda region.

2) New York-India. This route has been covered by the Flying Endeavor of American Export-Isbrandtsen Lines and follows a great circle route from New York to the Cape of Good Hope, the African Coast to Durban, crosses the Arabian Sea to several ports in India and Pakistan. It requires about 5 months for a complete circuit returning to New York, but provides coverage of seldom visited areas in the South Atlantic and Indian Oceans.

3) New York-Africa. This route follows a great circle from New York to Dakar or Freetown, and then proceeds to several African ports. When a southerly great circle route is followed returning from the Cape of Good Hope, a large area of extremely low aerosol concentrations is often encountered. It has been covered by the African Crescent of Farrell Lines, and requires about 90 days.



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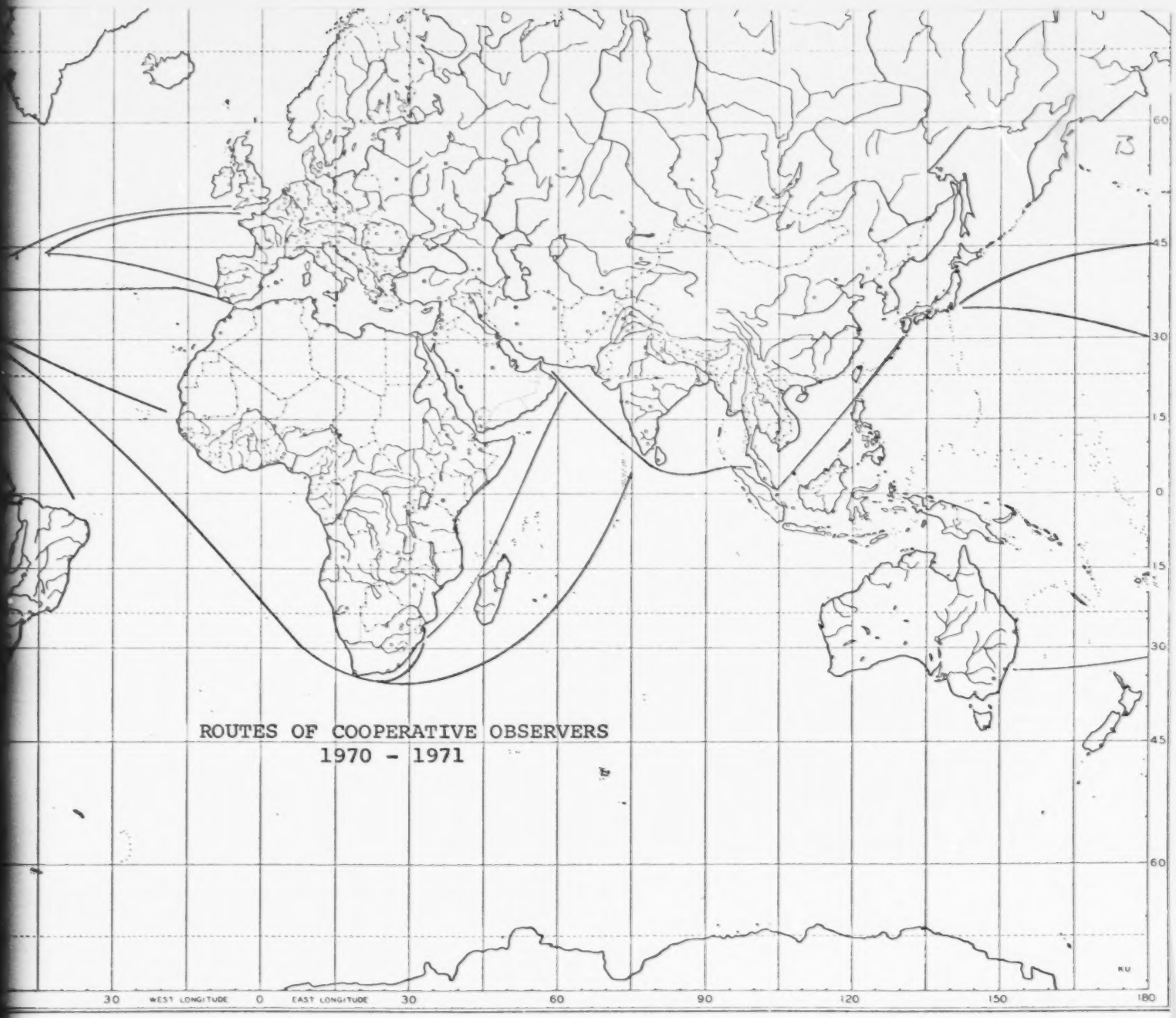
ROUTES OF COOPERATIVE OBSERVERS
1970 - 1971
60
45
30
15
0
15
30
45
60
30 WEST LONGITUDE 0 EAST LONGITUDE 30 60 90 120 150 180
Prepared by Henry M. Lepper
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FIGURE I

4) New York-Eastern South America. This route has been reported by the Moor Mac Cape, of the Moore McCormick Line. It traverses the Bermuda High area, where continental aerosol often exists several hundred miles off shore. A complete voyage requires about 60 days.

5) New York-Panama Canal-San Francisco-Honolulu-Yokohama. This route is covered by the American Legacy of U. S. Lines. It traverses the Bermuda High enroute to the Panama Canal and crosses the Pacific at low latitude from San Francisco-Hawaii and Hawaii-Yokohama. Five to six crossings per year can be made by this ship.

6) New York-Panama Canal-Australia. The Austral Patriot of Farrell Lines follows this route, requiring about 10-12 weeks to complete its voyage. It is an important route because it crosses a very rarely traveled portion of the Pacific in the vicinity of Pitcairn Island.

7) New York-Yokohama-Kuwait-New York. This unique around the world route is covered by the Mobil Energy, of Mobil Tankers Ltd. It covers the North Pacific on a great circle route from the Panama Canal to Yokohama or Kobe, and provides coverage of the Western Pacific and Indian Ocean when proceeding from Japan through the Malacca Straits to Kuwait.

8) New York-Bermuda-Azores-Gibraltar. This route and return is covered twice annually by the Maritime College training ship Empire State during training cruises. It traverses the Bermuda High-Azores High area, where continental aerosol often extends several hundred miles offshore.

9) Lisbon Portugal-Ocean Station Delta-Hamburg Germany. This unique sea route was followed by the Research Vessel, Meteor, as part of a Mediterranean Outflow Experiment. The principal investigator accompanied this expedition and was able to make aerosol observations in an area of the North Atlantic parallel to the trade routes.

The coverage obtained during this year has been most complete on the North Atlantic, and has produced sufficient data to calculate average values in many areas which are reported in Section VI. Data from the Pacific and Indian Oceans is more sparse, as only a few crossings have been completed in those areas, between the time of instrumentation availability and the preparation of this report. It is anticipated that additional instrumentation will be placed in Pacific and Indian Ocean service.

III INSTRUMENTATION

The instrumentation used in the cooperative program to date has been the portable Gardner Small Particle Detector, Type CN originally described by Rich (1955). A prototype photographic Aitken Counter (described in appendix "A") was also used during the Meteor Expedition.

The portable counter gives good service, and is well adapted to such cooperative ventures where little or no maintenance or spare parts are available on a routine basis. Each counter placed aboard ship is accompanied by a spare battery and pump as these are the components most likely to fail within one year.

Experience has shown that, after about one year's service at sea, it is desirable to return the instrument to the laboratory for servicing, as salt water corrosion, and internal buildup of salt crystals, begin to degrade instrument performance. With the exception of carrying case degradation, no ther deterioration seems to take place. Since it is desirable to recalibrate at annual intervals, this amount of degradation poses no additional logistic problems.

While this instrument has proven reliable at sea, the resolution of aerosol concentrations by photoelectric counters becomes progressively poorer as the aerosol concentration decreases. This is not a great handicap in areas near the continents where concentrations higher than 500 particles/cm^3 are encountered, but may be so if one wishes to be quantitative with aerosol concentrations measured in mid ocean, and after storms, when concentrations of less than 300 $^n/cm^3$ are found.

Because of this lack of resolution at low values, aerosol concentrations will be mapped by broad categories; i. e. 300 $^n/cm^3$, 300-500 $^n/cm^3$, 500 to 1000 $^n/cm^3$. This averts many of the resolution problems, except that of "zero", as the portable photoelectric counter has a rather non uniform response to concentrations below 200 $^n/cm^3$. When large areas of less than 200 $^n/cm^3$ aerosol concentrations are encountered, additional instrument development will be necessary, or a photographic Aitken counter incorporated into the program.

While great discrepancies were found between concentration values obtained by the several types of Aitken Counters at the recent Ft. Collins Workshop on Aerosol Instruments, reasonably good agreement was present at the lower values of concentration, which are of prime interest to this program. This program will report numbers for aerosol concentration as referred to a Pollak (1957) photoelectric counter, until some more suitable reference standard becomes available.

IV QUALITY AND RELIABILITY OF DATA

The instrumentation used for the detection and counting of aerosol particles in this program is well suited to such an experiment with many observers, as it is not greatly influenced by operator technique if reasonable care is taken. The greatest variable in the quality of data is in the selection of the actual site on the ships' superstructure where the observation will be made. Generally, the upwind wing of the flying bridge is a satisfactory location; but this is not always in free air on faster ships or at low relative wind speeds.

Observers aboard container ships have reported difficulties in obtaining uncontaminated air on several occasions. In some cases, this is due to stack gas becoming entrained in an eddy circulation, and descending to the bridge; soot and the smell of the stack are often reported in these cases. In other cases, the contamination seems to arise from the bluff (almost cubic) aerodynamic shape of a fully loaded container ship, causing large pockets of slightly negative pressure, and entraining relatively contaminated ship air.

An experiment is being conducted on the Empire State during the July and August 1971 training cruise in an attempt to evaluate low speed shipboard aerodynamics, and develop a guide for sampling site location. Two observers, with two counters, simultaneously make observations; one observer at the bow, or the upwind wing of the bridge, while the other observer moves about the ships' superstructure. These experiments are being repeated with several relative winds, to determine the locations of sites receiving uncontaminated air under many conditions.

The principal investigator had several experiences while making observations aboard the R. V. Meteor (see Appendix C) which indicated that uncontaminated air could be obtained under most conditions by simply extending a tube above and upwind of, the furthest projection of the bridge. Uncontaminated air could be obtained in this way even when galley and smoking smells could be detected by the observer only a few centimeters behind the rail. A small probe, constructed of tubing and a clamp, to reach upwind of the superstructure, will now be supplied to all cooperating ships as they arrive at home port.

V RECORDING, TRANSMITTAL AND STORAGE OF DATA

The aerosol data are recorded in the "additional message groups" column of a carbon copy of the Ships Weather Observations form, at the time of observation. When the ship returns to New York, this form is forwarded to ASRC where the data are extracted, and entered, as a function of position, in several log books. The data sheets are then forwarded to the student observers at the Maritime College, where the aerosol concentrations encountered are plotted on weather map bases, showing wind direction, sky cover and precipitation at time of observation.

When sufficient data are collected in a given geographic area (2° Lat. x 2° Long. blocks have been used so far), they are examined for consistency. Occassionally obviously contaminated data must be rejected; the remaining data are averaged and the resulting average concentration coded, and entered, as a function of position on a large Mercator projection.

When sufficient data are collected, they are grouped, as a function of local time, wind, sun and other meteorological conditions for analysis similar to that applied to the Meteor data in the appendix. This analysis will be done by the student observers at SUMC, using the Maritime College Computer Facilities.

VI PRELIMINARY DATA - ICE NUCLEUS OBSERVATIONS

Background concentrations of ice crystal forming nuclei were measured over the North Atlantic as part of the aerosol observation program during the June-July 1970 training cruise of the "Empire State". Iodine vapor was added to the air samples obtained during the July-August 1970 training period, and lead aerosols were detected as ice nuclei, using the Schaefer (1966) method.

The ice crystal forming nuclei were detected in a cold box-mixing chamber, constructed from a "Del Monico" home freezer, of about 0.5 m^3 volume, lined with black cloth in the manner described by Schaefer (1949). This unit is especially well suited to shipboard use as a mixing chamber because of its small size and undivided interior. It is very quiet in operation and is capable of cooling to -36° to -40°C. It can be regulated to maintain temperatures of -20° to -30° quite easily and can be swept over a wide temperature range by allowing the chamber to rise to near freezing with the compressor off, and then using the "fast freeze" feature to lower the temperature through the desired range.

Illumination was provided by an American Optical microscope illuminator. This is a very convenient light source, as it has a bright beam of variable width. The width of the beam is adjusted so that one (1) litre of the mixing chamber is illuminated to provide quantitative counting of the ice crystals formed on the nuclei.

The number of ice forming nuclei are measured by collecting a large (about $1M^3$) plastic bag of ambient air from the bow, or flying bridge, of the ship. An Aitken count is simultaneously taken to ensure that the bag sample is not contaminated. The bag is then quickly transferred to the meteorology lab, and the cold chamber is flushed with the contents of the bag. This flushing is repeated several times. If necessary, moisture is added to the chamber by exhaling gently along the wall, to form a cloud; often, sufficient moisture is present in the ambient air to form a cloud at chamber temperature. A few seconds are allowed for the ice crystals to grow to visible size, and the number of crystals in the light beam is determined. If fewer than 1 crystal/litre is present, the light beam is swept, and the number of crystals in the box determined. Upon completion of counting, the box is closed and allowed to clear itself of aerosols by diffusion and sedimentation. This clearing process enables two (2) observations to be made daily, without danger of cross contamination. Iodine vapor is added

to the chamber when lead detection is to be attempted. Because of the extreme difficulty in removing iodine contamination, all background experiments were completed before iodine was added to the chamber.

Background ice forming nucleus concentrations were found to be on the order of 1 per litre at -15^{o} to -20^{o}C, 1 to 5 per litre at -20^{o} to -25^{o} C, and sometimes greater than 10 per litre at temperatures lower than -25^{o}C, in the North Sea and in close proximity to the continents. Concentrations of 1 per litre or less were found in observations made at temperatures of -22^{o} to -26^{o}C while well off shore.

When iodine was added to the chamber, from 0 to 5 ice crystals per litre were detected at -20^{o} to -25^{o}C in mid ocean, and high numbers were detected near shore. These experiments are being repeated during the 1971 training cruises.

TOTAL AEROSOL (AITKEN NUCLEI) CONCENTRATIONS

The great bulk of the aerosol observations so far performed have been made during cruises of the Empire State. As these are training cruises, the ship crosses the Atlantic at a latitude of 38^{o} to 40^{o}N, where fine weather is usually encountered in the Bermuda-Azores High pressure region, facilitating boat drills and other seamanship training. As cooperative aerosol experiments on this route began in 1966, a small backlog of data is available. Our first experiments (Hogan et al, 1967) showed that a "continental field" that is, an area of relatively high aerosol concentration, exists for several hundred miles off the North American and European Coasts at this latitude.

Repeated experiments have shown that, on the average, the "continental field" extends about 600 miles west of the Iberian Peninsula, but, in situations of strong westerly flow, the field becomes very narrow. The continental field east of North America is rather extensive and is quite regularly found. The Aitken nucleus concentrations observed on four departures and approaches of New York Harbor, via Montauk, are plotted in Figure II. Analysis of this figure shows that continental aerosol is almost always found west of 67.5^{o}W, but that these high concentrations are rarely found east of 65^{o}W longitude. The rate of decrease in aerosol concentration, down to the 1000 $^{n}/cm^{3}$ level, is remarkably constant, indicating that this decrease is probably the result of dilution; ie. the spreading of a line source into a greater volume. In one case (the departure of 6-7 Jun) an extremely low (350 $^{n}/cm^{3}$) concentration was observed at 68.7^{o} longitude, concurrent with southerly winds and rain showers. The observations made four hours later after precipitation had ceased, and those following, are nearer to, but still below average, even though the wind had shifted to a more

landward direction. The lower values obtained during this period are probably the result of removal of particles by the precipitation encountered, in addition to a longer wind trajectory to land. These four repetitive series of observations lead to a conclusion that a persistent continental field is present to the east of the New York metropolitan area, although its strength and range can be diminished by precipitation, or southerly surface winds. As this field diminishes, and observed values of aerosol approach maritime levels between 65°W and 60°W longitude, 60°W has been chosen as a dividing line for analysis of maritime air, near this latitude in the Atlantic.

Data obtained between 60°W and 20°W at 38° to 40°N latitude during the four crossings made in 1970 have been analyzed with respect to the frequency with which concentrations were observed:

Concentration, $^{n}/cm^{3}$	<200	<300	<400	<500	<600	<800	<1000	<1200
Relative frequency of observation	2.3%	28.6%	41.1%	70.6%	81.3%	95.8%	99.2%	100%

Plotting this frequency distribution on probability paper yields a median concentration of 410 $^{n}/cm^{3}$ and shows the distribution of values to be very nearly log normal. When these data are compared with observations made in previous years

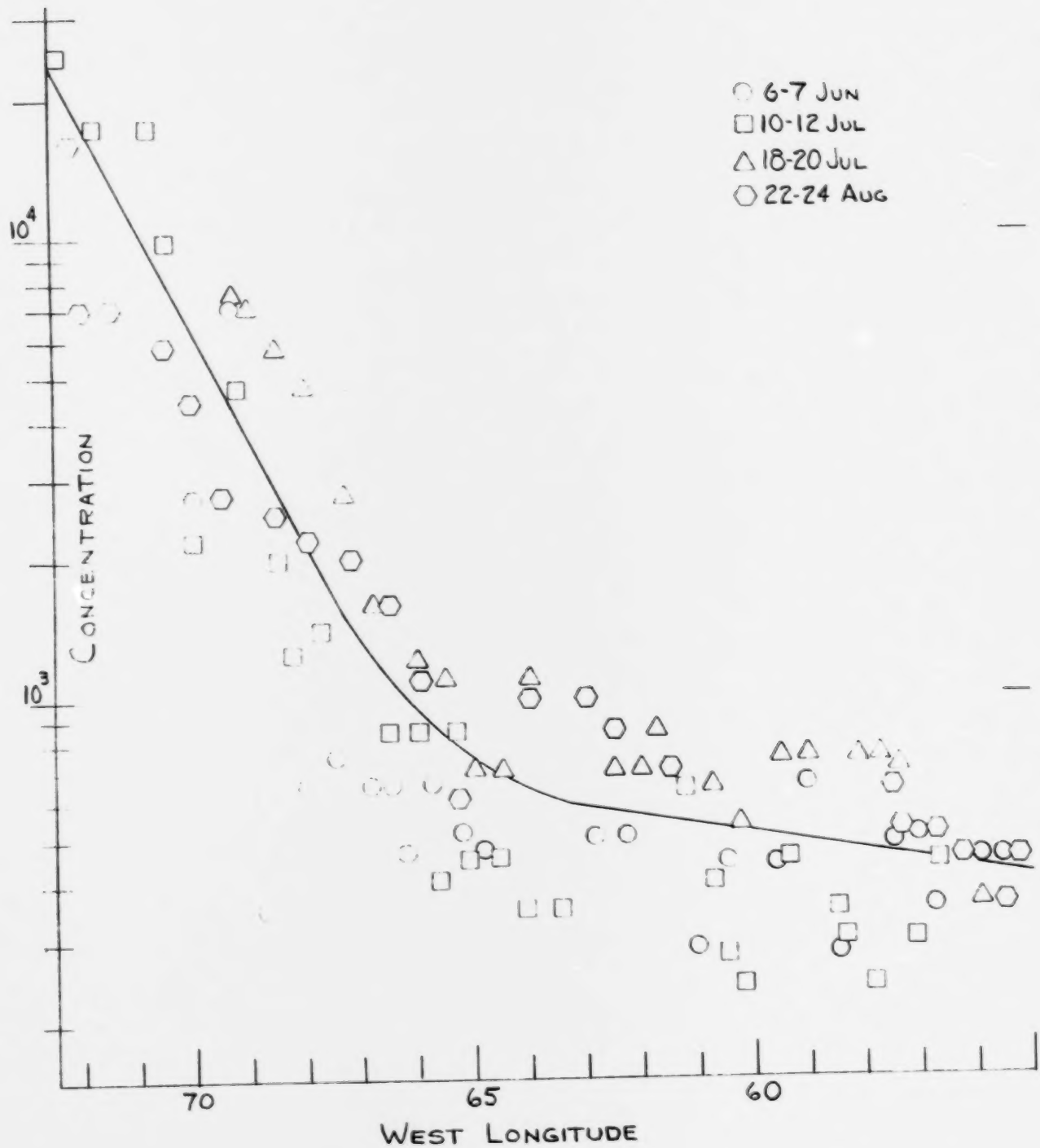
Year	16%	50%	84%	frequency less than level
1966	250 $^{n}/cm^{3}$	520 $^{n}/cm^{3}$	1050 $^{n}/cm^{3}$	
1968	280	580	1150	
1970	275	410	600	

the median value is apparently lower; the data are not directly comparable, however, as some more westerly data was included in the 1966 and 1968 analysis.

The data obtained during the four crossings made during summer, 1970, in the selected maritime area, were separated, and a frequency analysis performed.

		CONCENTRATIONS, N/CM^{3}							
Inclusive dates		<200	<300	<400	<500	<600	<800	<1000	<1200
6 Jun	14 Jun	1.5%	25.4	47.7	62.6	77.6	95.5	100	
4 Jul	10 Jul	0	19.1	35.0	58.7	74.6	93.6	96.8	100
19 Jul	26 Jul	6.9	50.0	63.8	77.8	84.6	95.8	100	
16 Aug	22 Aug	0	16.7	43.3	83.4	88.3	98.3	100	

FIGURE
AEROSOL CONCENTRATIONS
NEAR NORTH AMERICAN COAST
SUMMER 1970



Inclusive dates	16%	50%	84%	Percent Observations less than $^n/cm^3$
6 Jun 14 Jun	260	400	620	
4 Jul 10 Jul	300	460	700	
19 Jul 26 Jul	230	300	540	
16 Aug 22 Aug	300	530	590	

It is interesting to note that the widest departures from the median concentration, ($410\ ^n/cm^3$), for the entire period occurred during two adjacent periods which, in total, covered a time span of only 22 days. The number of relatively high readings did not change greatly over this period; but the number of relatively low ($<300\ ^n/cm^3$) values varied considerably. Concentrations less than $200\ ^n/cm^3$, which are relatively common in more northerly, and more southerly regions of the Atlantic, are very rarely found at 40°N, latitude.

Data obtained in previous programs have been combined with data from the first few months of this program, and average concentrations of Aitken particles occurring over the Atlantic Ocean, between the equator and 60°N latitude have been computed. The average values obtained in 2° Lat. x 2° Long. blocks have been entered on a Mercator projection (Figure III) and isopleths of equal nucleus concentration have been faired. Data have been reported from 54% of the area shown on the map; in some cases, only one or two observations have been received from the data block; in others 10 or more. These data are of course preliminary and subject to change, as the variability reported in the 40°N region shows, but they do present an insight to the areas where continental aerosol intrudes into the maritime regime, and to areas where essentially unmodified maritime aerosol exists. Additional data, recently observed along the New York - Cape of Good Hope - Great Circle route, indicate that a region of low ($<300\ ^n/cm^3$) aerosol concentrations is present in mid ocean south of 20°N latitude; data obtained during Meteor Expedition 23 (appendix C) verify the existence of the large region of low concentration north of 40°N latitude.

As most of the observations averaged to obtain the isopleths in Figure III was obtained during summer months, the plotted contours were compared to several meteorological variables plotted for these months in "the Marine Climatic Atlas of the World." While no readily apparent correlation exists, the band of relatively high (for oceanic air) concentration which separates the zones of low concentration in mid Atlantic coincides quite closely with the average 1025 mb isobar of summer high pressure. Analysis of this data, and the more recent data, with respect to correlation with marine meteorological data, is continuing at the Maritime College.

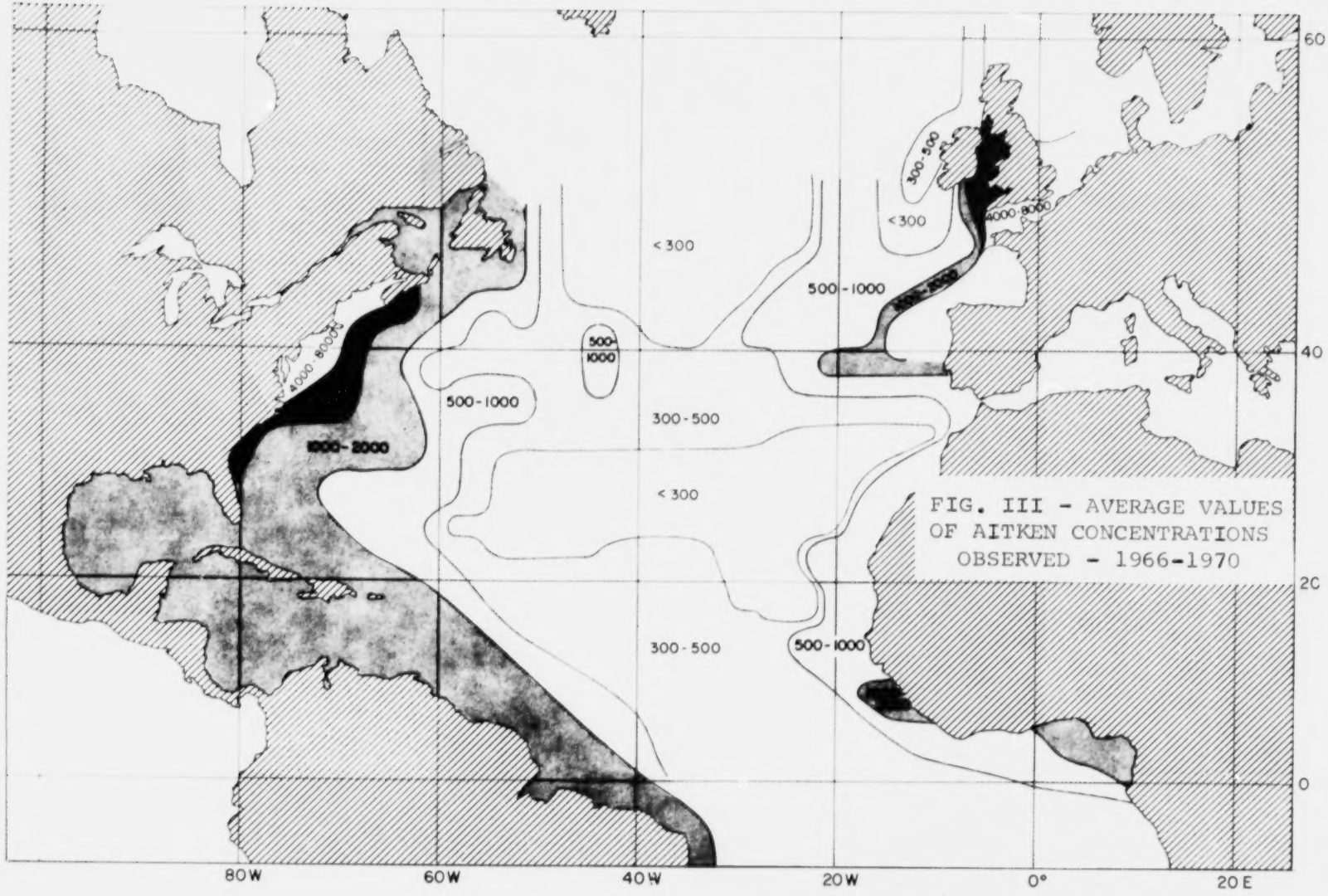


FIG. III - AVERAGE VALUES OF AITKEN CONCENTRATIONS OBSERVED - 1966-1970

OBSERVATIONS IN THE ALEUTIANS.

Several observations were made on the island of Shemya, during September, October and November 1970, through the cooperation of 1st Lt. T. G. Egelston, a former SUNYA student, then serving with the Air Weather Service. These observations were made on a daily basis, on the upwind shore of the island, and yielded fairly low values, the highest obtained being 995 $^{n}/cm^{3}$, 12% of all readings were below the threshold of the instrument. The median value of 420 $^{n}/cm^{3}$ obtained during the period is similar to that found at 40°N in the mid Atlantic.

Egelston also reported that, when the wind was from the direction of the islands of Attu (pop. 40 Coast Guardsmen) or Agattu (unpopulated) the concentration was higher than average. With wind from the open sea the average was lower.

CONTINENTAL EXPERIMENTS IN SUPPORT OF PROGRAM.

Two brief field experiments were conducted in support of the maritime observations of this program. The first, at the ASRC Whiteface Mountain Observatory on 24 and 25 Feb. 1971, was intended as a test of the aerosol detectors in conditions of dense cloud and icing.

When extremely low concentrations of aerosol are present, concurrent with fog or cloud droplets, photoelectric measurement of the aerosol concentration becomes difficult, due to light scatter in the expansion chamber from the droplets existing before the expansion takes place. This results in a zero error approximately equal to the number of cloud drops, which is not significant if the concentrations are simply grouped for frequency analysis, but is important if one wishes to find the minimum value of aerosol concentration. Also, if the instrument temperature is at or below ambient temperature when aspirated with air containing suspended fog drops, water will be condensed, and drops collected on the photocell, inducing larger errors. These effects can be minimized, but not eliminated, by maintaining the instrument at a slightly higher than ambient temperature. Stability of zero setting of the photoelectric counter can be used as a guide to ensure that, at least no photocell condensation is occurring. Storage of the instrument in the chart room, or other heated area, between observations, will minimize but not eliminate this problem at sea.

During the experiment period, the summit of the mountain and the observatory (elev. 4900' msl) were in clear skies, above the cloud deck, for two periods of several hours. The aerosol con-

centration fell to below 300 $^{n}/cm^{3}$ while the summit was above the cloud deck, but rose to several thousand (range 1150 $^{n}/cm^{3}$ to 5400 $^{n}/cm^{3}$ during period) when the summit was within the cloud, or below base. As the observatory is at an altitude to which mixing frequently occurs, and is surrounded by dense population and industrial centers, scavenging processes within the cloud must assist dilution processes to reduce the aerosol concentration to these levels.

A second field experiment was conducted in early March, in Nevada and California. Previous experiments (Hogan 1968) had shown that while unmodified maritime aerosol was not often found at the Pacific coast, it was often found atop the first ridge inland, during stormy periods. This experiment utilized our mobile laboratory, and aerosol equipment, to traverse a storm system across several ridges to determine if aerosol concentrations of a level usually associated with maritime air, can be detected far inland, during stormy periods.

Rather high (for unpopulated continental areas) aerosol concentrations of 3000 to 5000 $^{n}/cm^{3}$ were encountered under clear skies to $^{2}/10$ cirrus, while traveling from the ASRC Field Station at Flagstaff, Arizona, to the University of Nevada Field Station at Death Valley. Concentrations measured in a remote area in northern Death Valley under $^{3}/10$ cirrus to cirrus overcast on the 10th and 11th of March ranged from 700 to 1500 $^{n}/cm^{3}$, which is quite normal for the area (F. Went - private communication). At this time, a deep storm system was off the Pacific Northwest; measurements made in uninhabited valleys of central Nevada, later on the 11th, while still south of the surface front, but under cirrus and wave clouds associated with the storm, were from 380 to 460 $^{n}/cm^{3}$. Measurements made in rain and high winds, in several rural, but populated, areas of California (along a line roughly from Placerville to Ft. Bragg) during the morning of 12 Mar, showed low values of 700 to 1500 $^{n}/cm^{3}$. The rain ceased upon reaching the coast, and similar values were measured at several points along the coast to Eureka.

A rarely travelled road was followed from Eureka to Susanville, through occassional drizzle and snow flurries. Aerosol concentrations in the maritime range (from 290 to 420 $^{n}/cm^{3}$) were found atop the several ridges. Upon descending into the valley near Red Bluff, readings under $^{5}/10$ Cumulus were in the 5000 $^{n}/cm^{3}$ range.

On 14 March, very low concentration of 290 $^{n}/cm^{3}$ were measured north of Pyramid Lake, Nevada. While no precipitation reached the ground at the point of observation, snow was observed at Susanville,

and virga was observed beneath the overcast while making the aerosol measurements. Concentrations of 2600 to 4200 $^{n}/cm^{3}$ were measured in the same areas of central Nevada, while returning to the Flagstaff Field Station under varying cirrus cover, on 15 and 16 March, in apparently continental air.

While it is difficult to interpret data based on single observations at several points, it appears that unmodified, or only slightly modified Pacific aerosol can be found atop mountain ridges far inland from the Pacific coast. Additionally it appears that the scavenging action of these marine storm systems is sufficient to produce relatively low aerosol levels far inland, and even in populated areas. It is especially interesting to note that the number concentrations are reduced to very low levels at the edge of these storm systems, even though no precipitation reaches the ground.

VII CONCLUSIONS AND RECOMMENDATIONS

1) A cooperative shipboard observer network, where aerosol observations are performed concurrently with the cooperative synoptic meteorological observations, is a workable method for rapidly collecting aerosol data from oceanic areas.

2) The quality of the data collected to date is generally good; contamination problems have been present with some relative winds, on container ships, but this may be eliminated by a simple clamp-on probe, extending outside the superstructure. In general, contamination problems seem to increase with the speed of the ship.

3) Aerosol concentrations, over the Atlantic, are as variable as any other meteorological phenomena. Any attempts to define a secular change in aerosol concentration over the seas should be based on frequent long term measurements of concentration.

4) A region of transition, from continentally modified aerosol, to maritime aerosol, has been found in the region surrounding $40^{o}N \times 65^{o}W$. Studies of atmospheric aerosol removal processes could be productively performed in that area.

5) A large area of apparently unmodified maritime aerosol, has been located in mid Atlantic between 45^{o} and $50^{o}N$ latitude, another such area appears to exist south of $30^{o}N$. An analysis of synoptic and satellite data for these areas might aid in understanding of natural processes which remove atmospheric particles.

6) The aerosol concentrations measured along $40^{o}N$ above the Atlantic are often higher than concentrations observed at similar latitudes in California and Nevada when Pacific air, with associated cloudiness and precipitation, is present. This suggests that, in this area of relatively stable high pressure, the absence of scavenging mechanisms may be a major factor in allowing this band of relatively high (for maritime air) concentrations of aerosols to span the Atlantic.

7) Experiments aboard the Meteor showed fog to be an exceptionally good scavenger for aerosol particles; continental stratus may then be an equally effective scavenger, producing low concentrations as those observed at Whiteface Observatory 24 Feb 1971.

SECTION VIII ACKNOWLEDGEMENT

The authors wish to thank the management, and port personnel, of the several shipping lines who cooperated in this observation program. Our special gratitude is extended to the Masters of the cooperating ships, and to the deck officers and King's Point Cadets who performed the many observations.

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APPENDIX A

A PORTABLE PHOTO RECORDING AITKEN NUCLEUS COUNTER

Introduction:

The commercial Gardner Associates Small Particle Detector (Rich 1955), replicas of the Pollak (1955) photo electric nucleus counter, Sholz, and Aitken Counters of several manufactures, have been used by many investigators to determine the atmospheric aerosol concentrations at remote maritime and continental sites. The results obtained with these instruments have generally been good, especially when the photo electric counters have been used in continentally contaminated air containing aerosol concentrations of 10^3 to 10^5 particles/cm^3.

When concentrations of 10^2 to 10^3 particles/cm^3 are encountered, as is often the case in offshore maritime air, the sensitivity of the photo electric counters is such that is is difficult to resolve concentrations to better than $\pm$100 particles/cm^3 in the range of 200 to 1000 particles/cm^3, and the instrument responds erratically to concentrations below 200 particles/cm^3. The Aitken and Sholz type counters would appear to be superior when low concentrations are encountered; as the individual drops are counted, and the concentration geometrically calculated. This counting procedure becomes complicated at low concentrations, as the counting error becomes progressively larger with decreasing concentration, requiring a large number of counts to be made to accurately measure the existing number concentration of aerosol particles. These repeated measurements are acceptable if the observer is assigned as a full time aerosol observer, and only the approximate number concentration of aerosol particles is desired. If, however, the observer has other observations to perform, and/or accurate aerosol counts are desired, as is necessitated by sizing techniques which are based on the ratio of aerosol concentrations transmitted by a sizing device, then the Aitken and Sholz counters become too cumbersome. Additionally the dilution systems required by the Aitken and Sholz counters, and the manual methods of forming the Aitken expansion, lead to uncertainties when a large number of instruments are operated by several individuals.

A photographic recording Aitken Counter has been developed for field and maritime use, with the original design philosophy based on the following criterion:

1) Automatic operation of the expansion sequence, to minimize differences in operator technique.

2) Photographic recording, with capability for multiple expansions and exposures on a single film sheet to minimize random counting error.

3) Self processing Polaroid® film to be used in recording, to provide rapid access to data, and also to reduce coding and bookkeeping errors by allowing the observer to record location, data, time and number of expansions directly on the frame immediately upon performing the measurement.

4) "Absolute" counting of a thin illuminated volume, which allows a purely geometric calibration of the instrument by measuring the dimensions of the light beam.

5) Battery operation of all circuits, small size, and light weight to facilitate remote site operation.

6) Relatively low cost to permit a moderately dense network of observers.

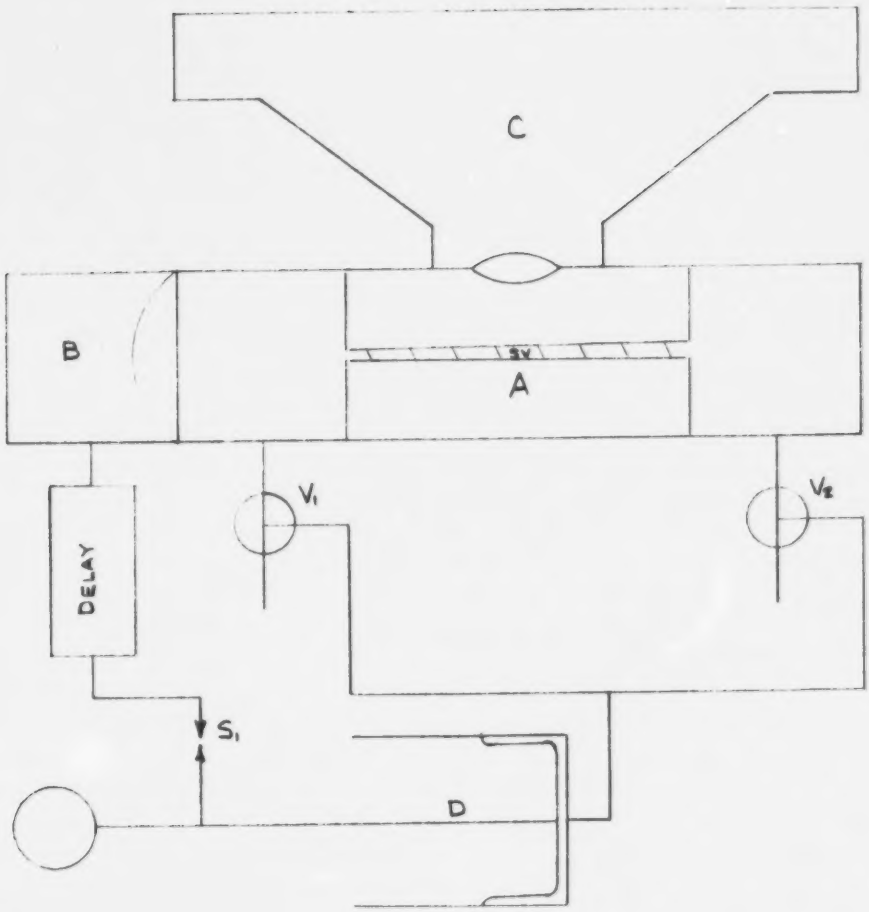
Two prototype instruments have been constructed, compared with photo electric counters, and field tested in remote continental and maritime areas.

Description of Instrument:

The instrument is shown schematically in Figure I, and consists of four (4) major components; A, a cloud chamber lined with water saturated wicking; B, a photo flash lamp; C, a Polaroid® camera back, lens and shutter assembly, and D, a Bellofram® piston and cylinder assembly. The cloud chamber of the prototype instrument is rectangular 3.6 x 3.8 x 1.9 cm inside dimensions, and is connected to the Bellofram® piston and cylinder through two (2) three way valves; the Aitken expansion is attained by releasing the spring loaded Bellofram® piston, and allowing the differential pressure across the piston to come into equilibrium with the spring force. This ensures a constant differential pressure, and a uniform rate of attaining the final pressure, without consideration of operator technique.

The operating cycle of the instrument is best described by referring to Figure I as a guide. The cloud chamber is aspirated by placing valves V_1 and V_2 in the position shown in the schematic and pulling atmospheric air through the chamber with a small pump, until the chamber has been flushed with 5 to 10 times its own volume of ambient air. Valves V_1 and V_2 are then rotated 90°, isolating the chamber from ambient air, but maintaining connection with the Bellofram® piston and cylinder. The oscillator charging the capacitor for the flash lamp ("B") is turned on, and the cloud chamber is allowed to come to temperature and vapor pressure equilibrium with the wet walls, while the capacitor is charging from the internal battery.

FIGURE I



OPERATIONAL SCHEMATIC

(1) VALVES (V_1) AND (V_2) ARE OPENED AS SHOWN, AND THE CLOUD CHAMBER (A) IS ASPIRATED.

(2) VALVES (V_1) AND (V_2) ARE ROTATED 90°, AND THE CHAMBER ALLOWED TO COME TO TEMPERATURE AND HUMIDITY EQUILIBRIUM.

(3) THE SPRING LOADED BELLOFRAM (D) IS RELEASED TO INITIATE THE AITKEN EXPANSION: SIMULTANEOUSLY, THE MAGNETIC SWITCH (S_1) INITIATES AN ELECTRICAL IMPULSE THROUGH A TIME DELAY

(4) THE CLOUD DROPS ARE ALLOWED TO GROW FOR 400 M SEC AFTER INITIATION OF THE EXPANSION; AT THIS TIME THE TIME DELAY TRIGGERS THE FLASH LAMP (B), WHICH ILLUMINATES THE SENSITIVE VOLUME (SV), RECORDING THE DROP IMAGES ON THE PHOTOGRAPHIC FILM IN THE RECORDING CAMERA, (C).

(5) VALVE (V_1) IS ROTATED 90°, THE BELLOFRAM RECOCKED, (V_2) ROTATED 90° AND THE INSTRUMENT ASPIRATED FOR THE NEXT OBSERVATION.

The camera shutter is opened, and the Bellofram® latch is released, when a small indicator light shows the capacitor is fully charged. Upon releasing the latch, the spring loaded Bellofram® begins the expansion, simultaneously activating a magnetic switch, which initiates a signal through a time delay circuit. The bellofram® rapidly comes to equilibrium at the Aitken differential pressure, as determined by the spring constant; 400 milliseconds after the expansion was initiated the time delay circuit fires the photoflash unit, illuminating the cloud drops in a narrow ribbon within the cloud chamber, and the images are recorded on the photographic film. The shutter is then closed, the system returned to ambient pressure by returning V1 to its original position and re-cocking and latching the bellofram® piston.

The number of cloud drops are now recorded on the photographic film; it can be immediately processed and examined, or the entire cycle can be repeated one or more times to reduce the random counting error when small concentrations are present.

The cycle described above can be accomplished in as little as 20 seconds; however, it appears that the most repeatable and reliable data can be obtained by slowly flushing the instrument and allowing ample time for the air sample to come to temperature equilibrium; this "slow" cycle requires about 60 seconds to perform.

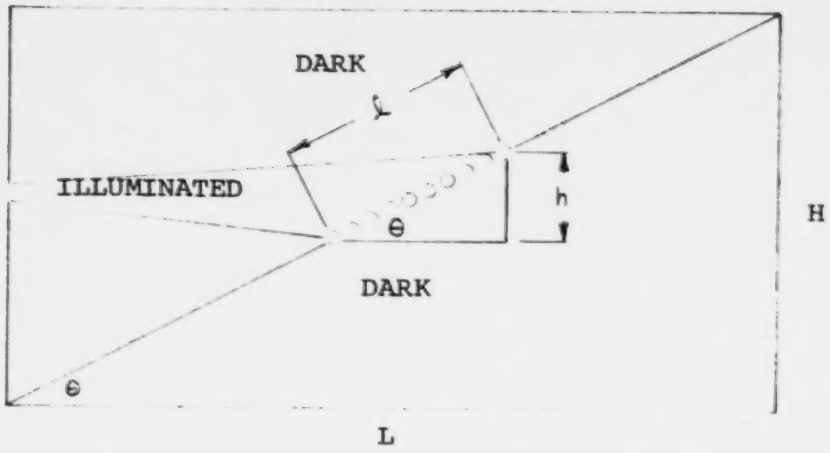
Calibration of Instrument:

This instrument is, in theory, an "Absolute Aitken Counter," that is, its calibration depends only on the precise measurement of the ratio of the volume of the illuminated zone to the area of the photograph, and the verification that the Aitken expansion of P (original) = 1.21 P (final) is attained. The spring loaded bellofram® piston achieves the Aitken supersaturation with great repeatability, and is easily measured with an Absolute Pressure gauge.

The optical magnification of the lens system is measured by placing a ruled grid of 1 mm squares in the illuminated volume of the cloud chamber, reassembling the chamber and camera system, and photographing the grid, using neutral density filters to attenuate the bright light source used to illuminate the cloud drops. The size of the image of the 1mm grids is then measured under a low power microscope and the linear and areal magnification calculated.

The width of the light beam, and consequently the depth of illuminated volume, is determined by placing (as shown in Fig. II) a rigidly supported section of 100 mesh (40 wires to 1 cm) brass filter screen at an angle of 27.3° in the cloud chamber. The mesh

FIGURE II



CALIBRATION OF SENSITIVE VOLUME

(1) LENGTH (L) AND HEIGHT (H) ARE MEASURED.

(2) RIGID SUPPORT OF LENGTH $\sqrt{L^2 + H^2}$ IS PREPARED, AND A SECTION OF 100 MESH SCREEN MOUNTED.

(3) ILLUMINATION ENTERING THROUGH SLIT AT LEFT IS ATTENUATED WITH 1% + 30% N. D. FILTERS.

(4) SCREEN IS INSERTED IN CHAMBER AT GEOMETRICALLY DETERMINED ANGLE θ, CHAMBER AND CAMERA REASSEMBLED.

(5) SEVERAL PHOTOGRAPHS ARE TAKEN, AND AVERAGE NUMBER OF WIRES ILLUMINATED DETERMINED.

(6) AS (ℓ) IS A MULTIPLE OF THE NUMBER OF WIRES, AND θ IS KNOWN, THE DEPTH ILLUMINATED (h) CAN BE DETERMINED TRIGONOMETRICALLY.

is photographed with the same camera and neutral density filter arrangement, and the number of wires counted with a low power microscope. The number of wires counted, times their spacing of the wires, is then equal to the hypotenuse of a right triangle; since the included angle of 27.3^{o} is known, the height of the triangle, which is the width (d) of the light beam, can be determined.

The product of the reciprocals of the optical gain and the volume illuminated define the multiplier to translate the number of drop images in 1 cm^2 of film to 1 cm^3 of fog in the chamber. Since the initial sample was expanded 1.21 times to obtain the cloud, the number of cloud drops is multiplied by 1.21 to obtain the original number of particles.

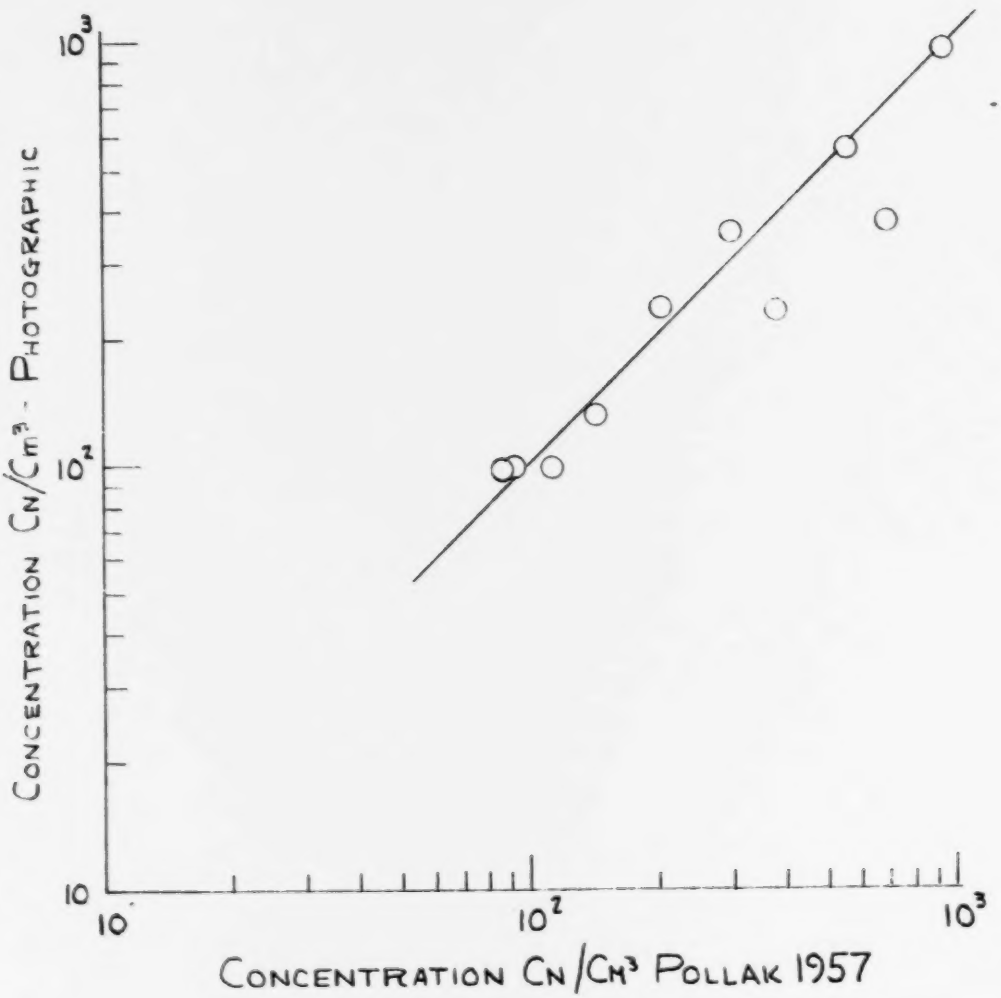
Comparison With Photo Electric Instruments

The second of the prototype instruments was compared with a Dublin Pollak 1957 photo electric nucleus counter, on dilute room air aerosol. Room air was mixed with filtered air, stored in a 200 litre metal drum, and continuously stirred. The container was continuously flushed with filtered air during the experiment, to produce a decreasing aerosol concentration for comparison purposes, from 1000 particles/cm^3 to 100 particles/cm^3. The result of the comparison are shown in Figure III, and with the exception of two "flyers" probably caused by incomplete flushing, are in quite good agreement. A more complete and exact comparison, with the "Absolute" counter of Junge, at the Max Planck Institute fur Chemie, is now in progress. The comparison with the Pollak counter, however, confirmed that the instrument is worthy of field trial, and further development.

Techniques of Droplet Counting

The droplet images formed on the recording film are not sharp, due to the simple lens used, and the amount of magnification. This is unimportant in the reduction of data, as the number of images formed is governed by the volume illuminated, and not by the depth of field of the lens. The images are of good contrast and are quite easy to see and count under low power magnification; however, small emulsion separations, and sedimenting dust also produce drop like spots on the polaroid film. The use of a low power stero microscope for counting drop images allows the analyst to uniquely separate drop images from film defects and dust particles, and allows many counts to be made without fatigue. The AO "Forty" student microscope is especially suited for this counting operation, as it has a built in diffuse light source at 60^{o} to the viewing plane which minimizes film reflections, and has a field of view of almost exactly 1 cm^2, which eliminates one calculation in the data reduction process.

FIGURE III
COMPARISON OF PHOTOGRAPHIC
AITKEN COUNTER WITH DUBLIN POLLAK



During the initial laboratory development and comparison of the prototype instrument, the film records were analyzed by ruling two mutually perpendicular lines through the densest field of drop images, and counting the number of drops in 1 cm^2 in each quadrant. Later, during field development, this technique was modified to scribing one centerline through the droplet field, with three perpendiculars and counting 8 cm^2. A matrix was constructed from 116 frames obtained in counting maritime aerosol over the North Atlantic in this manner. Examining the matrix shows that there is symetry and uniform distribution on the left and right sides of the axis,

307	260
483	553
608	615
541	520

but that the uppermost two (2) cm^2 of the matrix have consistently fewer droplets. This is probably due to poor contrast and resolution of this simple lens as one departs from the central area, but amounts to a systematic error of only 12% through the course of these measurements.

This analysis then shows that the best counting technique is to scribe a centerline, through the droplet field, and count the six (6) cm^2 surrounding the optical axis.

The low aerosol concentrations which this instrument was designed to measure result in records showing only one or two drop images per centimeter square when a single expansion and exposure is made on a single film sheet. As the counting error is large when only a few images are counted, it is desirable to increase the number of images counted per experiment. This is most easily accomplished by operating the instrument through several flush-expand-photograph cycles on a single frame of film, increasing the number of images to be counted, and substantially reduces the counting error.

This requires that the camera-cloud chamber interface is exceptionally light tight to prevent fogging of film by stray light while the shutter is open. A simple sponge rubber gasket achieved sufficient sealing to allow ten exposures without loss of contrast in the prototype, even in bright sunlight.

Field experiments show that three (3) or five (5) repetitive cycles produce a sufficient number of drop images to yield an accurate particle count in the range 100 to 1000 particles/cm^3 encountered over the sea. When extremely low (ie. $<$ 100 particles/cm^3) aerosol concentrations are present, 10 cycles may produce more satisfactory results.

Results of Field Tests

The prototype instruments were field tested in maritime air along the west coast of northern California, in dry continental air in Nevada and Arizona, and aboard ship in the North Atlantic. The aerosol concentrations measured with this and other instruments during these experiments are reported in the body of this report.

The expansion spring was adjusted to produce the Aitken pressure ratio of 1.21 to 1 at sea level; no adjustments were made during the field tests, but measurements of the pressure ratio were made at two other altitudes:

Location	Altitude, Msl	Po	Pf	Po/Pf
Flagstaff, Ariz.	6900'	23.35 In Hg	19.20 In Hg	1.215
Death Valley, Nev.	2000'	27.95	23.60	1.185
Flagstaff, Ariz.	6900'	23.45	19.20	1.22

While the instrument does not precisely achieve the Aitken supersaturation at all times and all altitudes it is sufficiently close to introduce minimal error in the counting results.

Several minor problems were encountered in field testing; (1) the relatively inexpensive film back leaked light in prolonged exposure to desert sun; (2) heavy rain penetrated the flash lamp, shorting the switch contacts until dried and (3) blowing oceanic drizzle often softened the film release tab, preventing processing of that frame. All of these problems can be overcome by enclosing the instrument in a carrying case.

Summary and Conclusions

A portable photo recording Aitken Counter has been developed for field aerosol experiments in remote areas where concentrations of less than 1000 particles/cm^3 are often encountered. This instrument if in good agreement with laboratory photo electric counters, and is undergoing calibration with reference to a more sophisticated photographic counter. The counter has proven reliable in field tests in remote continental areas, and aboard ship.

APPENDIX B

A FIELD INSTRUMENT FOR AEROSOL SIZE MEASUREMENT

Introduction

A combined diffusion battery - denuder, useful in obtaining size parameters of a real atmospheric aerosol, was described by Rich (1965). This instrument consists of 21 parallel metal plates, forming 20 narrow channels. When all plates are maintained at the same potential, the device acts as a conventional diffusion battery; when a potential is applied across alternate plates, the instrument becomes, both theoretically and practically, a diffusion battery and a denuder in series. A graphical technique is then used to determine two equivalent monodisperse sizes from the diffusion loss and the percent uncharged. Rich's original diffuser denuder was designed to operate at a flow of 6 litres/min in conjunction with a General Electric Continuous Nucleus Counter (Skala 1963). This combination was very useful in determining the size of urban or rural aerosol when operated at a field station or as part of a mobil laboratory, but was limited by its lack of portability and high power requirements, and consequently seldom applied to studies of remote continental, or maritime aerosols.

Remote area studies often utilize the Gardner-Rich photo electric counter, the Aitken or Sholz counter or the photo recording Aitken Counter described in appendix A. These studies would be greatly enhanced, if it were possible to include a measure of some size parameter of this aerosol, particularly if this size, even though an "equivalent" size, could be directly related to the sizes previously obtained with the Diffuser-Denuder/Continuous Nucleus Counter combination.

Description of Instrument

A single channel diffuser-denuder has been constructed, utilizing the Rich 1965 geometry, but only two parallel plates. As the original battery of twenty channels operated at 100 cm^3/sec, or 5 cm^3/sec/channel, the smaller instrument will produce an equivalent diffusion loss, and remove all charged particles at this same specific flow rate. This slow flow rate (5 cm^3/sec) is compatible with the valving of the several portable counters, and is easily produced for sufficiently long periods in the field by allowing water to flow from a litre container through a calibrated orifice. A brushless D. C. motor driven pump is also suitable as an air mover for this system at a slight sacrifice in weight. A single 12V lantern battery will provide sufficient power to operate these pumps at .300 lpm for well over 100 hours.

The unit is quite easily constructed from 0.030" sheet aluminum, and is well suited for manufacture of replicas by any small sheet metal shop. The prototype was constructed by the author in one day, and required only a drill press and band saw in addition to hand tools. The construction is greatly facilitated by the use of high resistance micarta or fibreglass stock of 0.793 mm (1/32 inch) thickness as spacers between the plates, and nylon screws to join all parts. A photograph is attached following the text. The original unit was stiffened by the addition of two pieces of ½" aluminum angle attached along the edges of the assembly, to provide rigidity when shipping the item in a suitcase or trunk to an expedition, or for portability in a briefcase. The dimensions of the diffuser-denuder are such that it may be attached to the case cover of the Gardner-Rich small particle detector.

Instructions for Use

The combination diffuser-denuder is used in conjunction with any aerosol detector capable of being operated at a flow of 5 cm^3/sec. The total concentration of aerosol (Zo) is first determined, using no hose or connecting tubing on the inlet of the detector. The diffusion battery is then inserted at the inlet of the detector, with the plates connected at common potential, and aspirated at 5 cm^3/sec. until the instrument volume is thoroughly flushed; the concentration at the outlet (Z_A) is then determined. A potential of at least 90 volts is then applied across the plates, the instrument thoroughly flushed at 5 cm^3/sec, and the concentration of uncharged particles (Z_B) is then determined. Rich's equivalent radii can then be determined from the percent transmission (Z_A/Zo) when the unit is used as a diffusion battery, and the fraction charged (Z_B/Z_A) when operated as a denuder, using the curves shown in Figure I, which are calculated from the equations describing the original 20 channel battery.

Connecting tubing should not be used ahead of the detector when Zo is determined, as small particles will be lost in the tubing due to diffusion, producing a change in the Z_A/Zo ratio that will result in an erroneously large equivalent diffusion size. If this tubing is non conducting, charged particles will also be removed, altering the Z_B/Z_A) fraction to make the equivalent aerosol appear to be smaller. It is permissible to use a short length of connecting tubing between the diffusion battery and the detector, but this tubing should be internally coated with glycerine if it is non-conducting.

Sea spray presents some problems in operation of a parallel plate device, as the salt crystals will bridge the air gap, shorting the potential, and will also corrode the aluminum. Protective coatings should not be used, as they will alter the local field,

and will enhance potential buildup when operating the device in the diffusion mode. The prototype was successfully operated for 21 days at sea. Flushing the instrument with fresh water whenever a salt buildup became noticeable eliminated damage to the instrument.

Summary and Conclusions

A portable diffuser-denuder, using the Rich 1965 geometry, and compatible with portable aerosol instrument has been developed. The instrument has been successfully used to determine the equivalent sizes of aerosol particles in maritime air; and is adaptable to measurement in any remote area where portability and reliability are prime considerations in instrument selection.

Reference - Rich, T. A., Apparatus and Method For Measuring the Size of Aerosols, Journal Des Recherches Atmospheriques Vol II, 2^{e} Annee Nos 2-3, Avril Septembre 1966.

FIGURE I

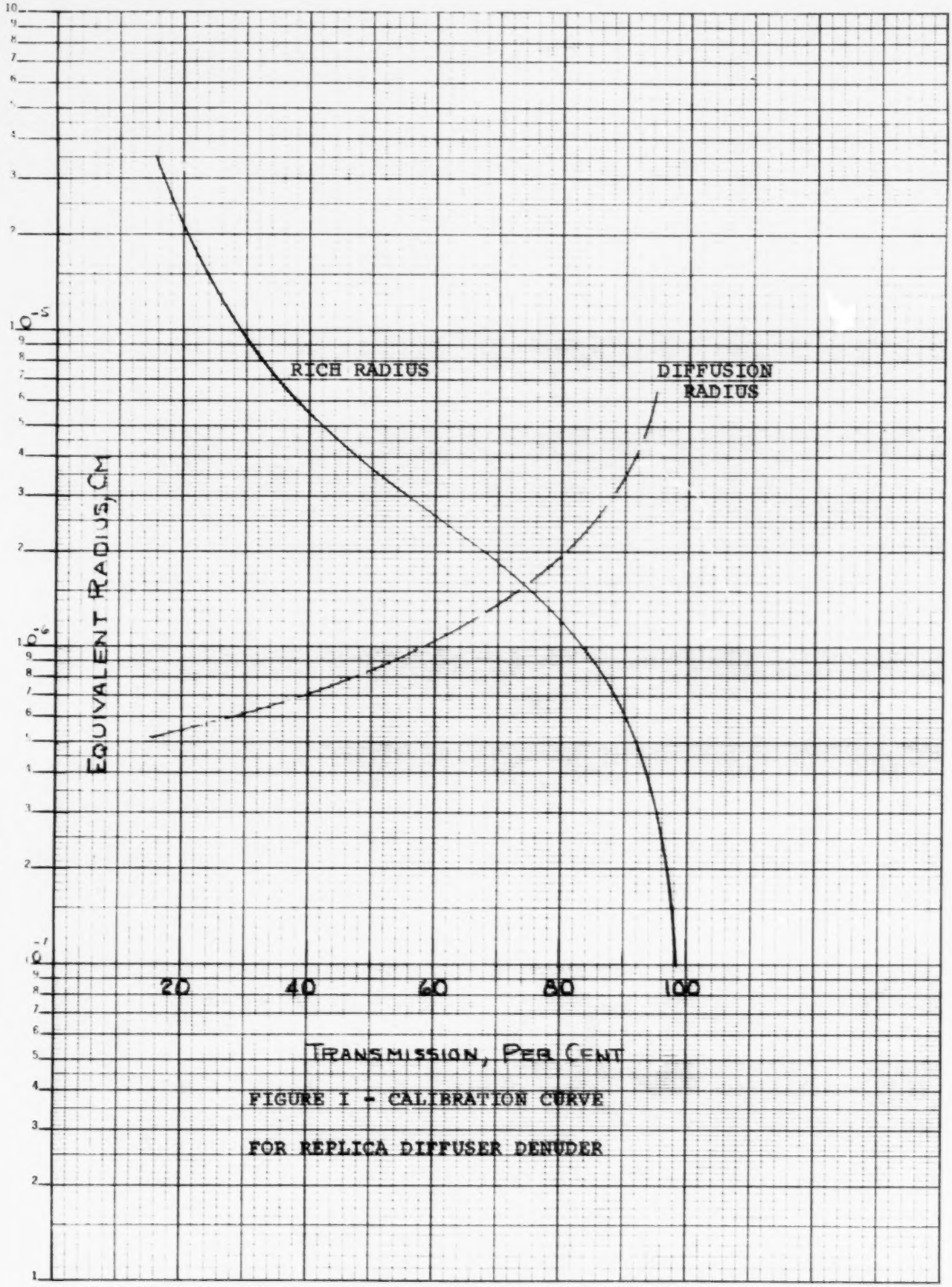
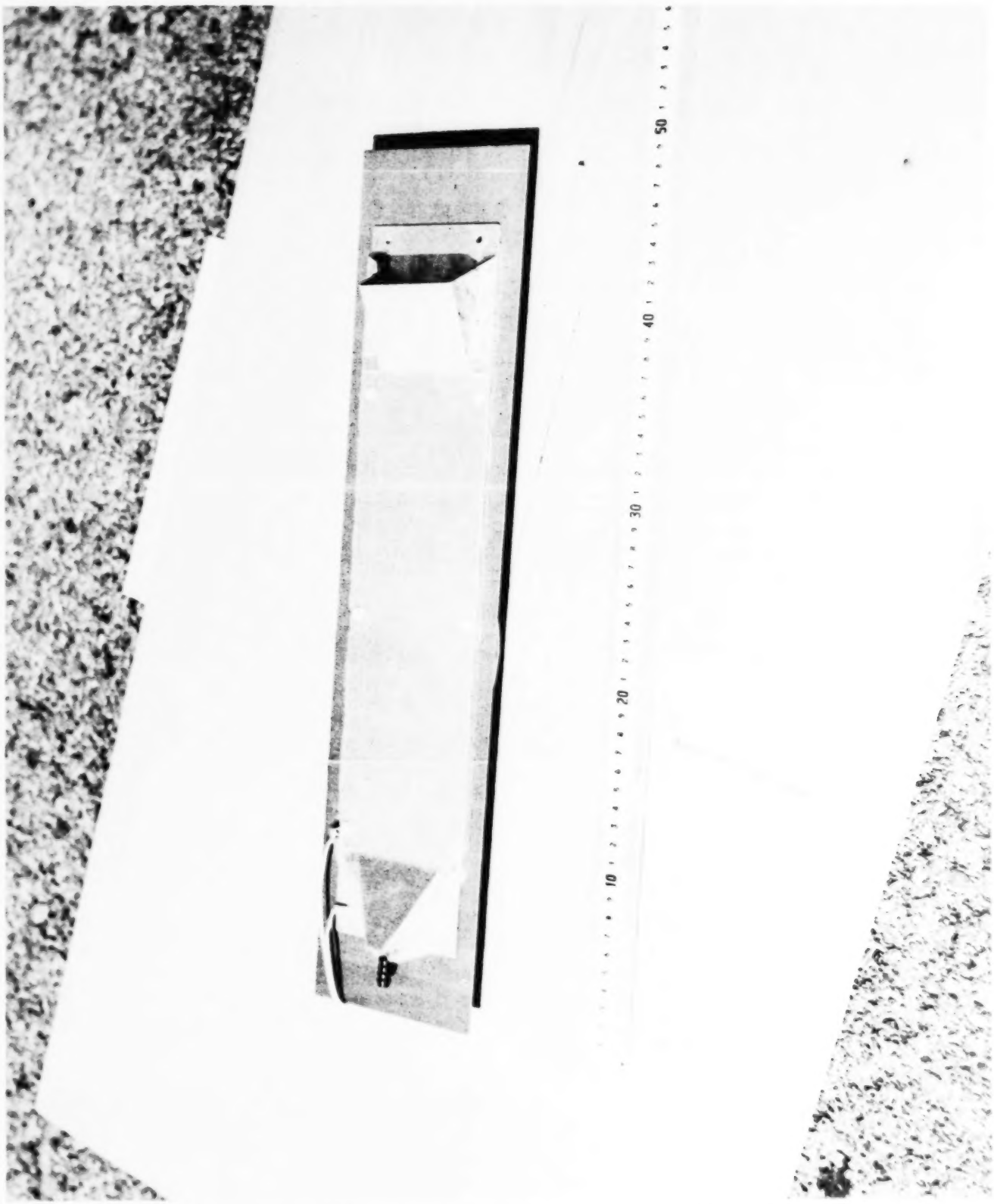


FIGURE I - CALIBRATION CURVE

FOR REPLICA DIFFUSER DENUDER



PHOTOGRAPH OF PORTABLE DIFFUSION BATTERY

APPENDIX C

RECENT AEROSOL OBSERVATIONS OVER THE NORTH ATLANTIC

Introduction

Aerosol observations were made, during Voyage 23 of the Research Vessel "Meteor," in an area of the North Atlantic Ocean lying approximately along a line connecting Lisbon, Portugal, Ocean Station Delta and the English Channel. This area is parallel to major shipping lanes, and is therefore of great interest because it presents an area of the North Atlantic which is almost entirely free of local contamination, and is not covered by a cooperative observer in the ASRC Maritime aerosol program.

The aerosol observations were made from the piel deck (i.e., uppermost deck, 16 meters above the water line) when the ship was under way, and from the piel deck or stern when the ship was dead in the water, on station. A small, movable table was used as an instrument stand on the piel deck. This table was located at the forward, port or starboard rail, depending on the relative wind. While it was quite difficult to obtain uncontaminated air inboard of the rail, the simple expedient of extending either the instrument, or a short length of tubing, about 50 cm above and ahead, or outboard of the rail would yield uncontaminated readings when the relative wind was from either forward quadrant. When the relative wind was from the aft quadrants, as was usually the case when the ship was on station, aerosol observations were made from the aftermost portion of the stern on D deck, about 4 meters above the water line.

When the relative wind velocity was small, it was necessary to move the observation site quite frequently, as outflow from ventilators could often be detected, as could the odors from tobacco smoking on the decks below. It would be desirable, on any future aerosol expedition, to extend three tubes of about 5-10 cm diameter, 3 meters forward, port and starboard of the highest deck, with a small suction fan pulling air down these tubes. This would ensure that all instruments receive uncontaminated air.

Experiments with the aerosol detectors on the piel deck show that, even in the case of a relatively slow moving ship such as the Meteor, a thin boundary layer of "ship air" is entrained along the vertical superstructure, and flows over the uppermost deck. Uncontaminated air can be found by reaching beyond this boundary layer with a short probe. A thicker boundary layer may be present on faster ships.

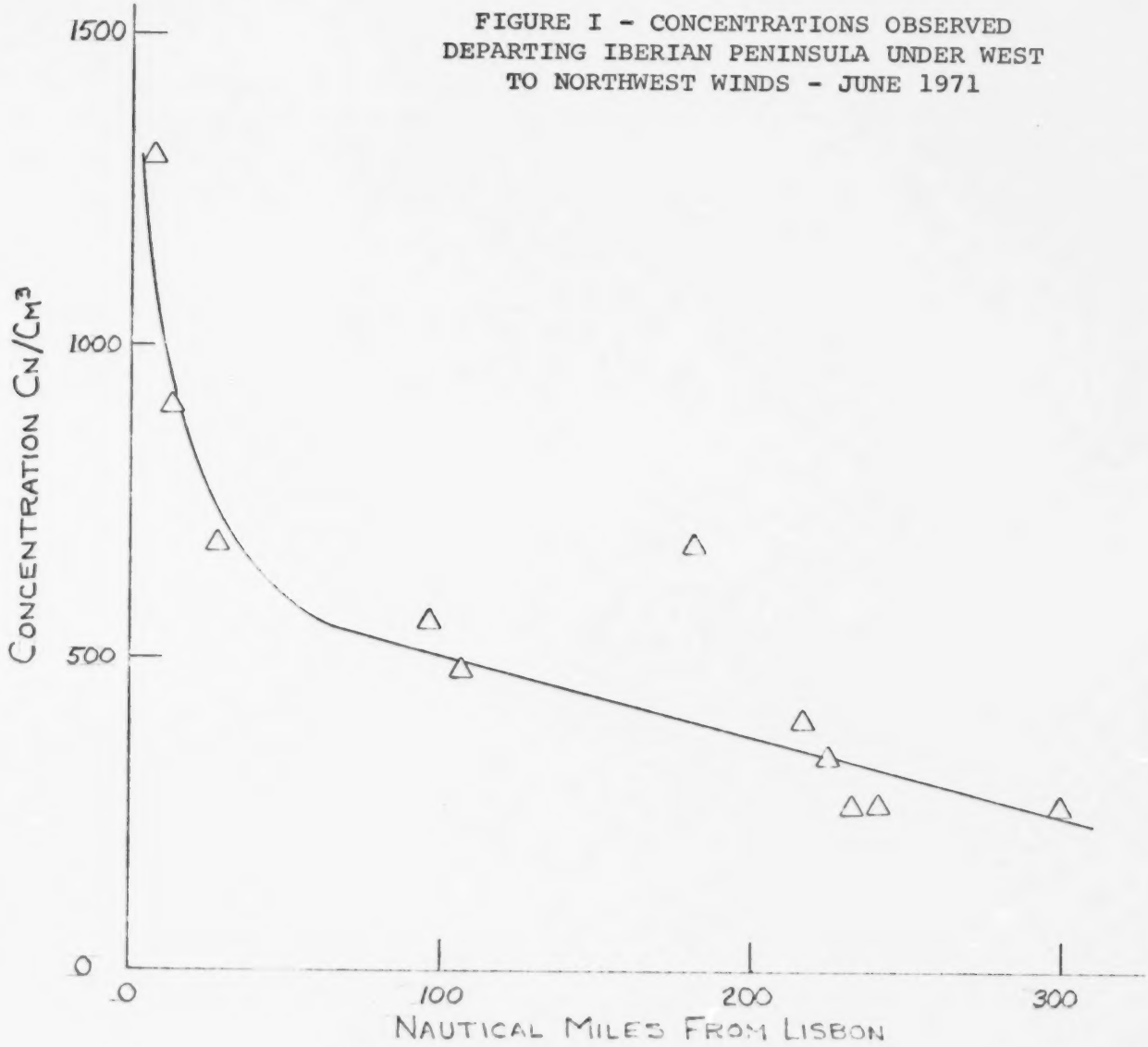
The aerosol detection instruments used, powered by internal batteries, posed little problem when it was necessary to move them about the ships' superstructure to obtain uncontaminated air. A photo electric nucleus counter was used to establish the relative aerosol level at each observation; when this level was below 500 particles/cm^3, in the region where resolution is poor with the photoelectric counter, a photographic Aitken Counter (described in appendix A) was used. A combined diffuser-denuder (described in appendix B) was used to determine the equivalent size of the aerosol several times during the voyage.

Sizes obtained were the Rich (1965) equivalent radii, determined by first measuring the total concentration (Z) of aerosol particles. A diffusion battery was then inserted ahead of the aerosol counter, and the concentration (De) transmitted through diffuser was determined. A potential of 90 volts was impressed across the plates of the diffuser, and the charged particles were removed; the concentration of uncharged particles (De_o) was then counted. The equivalent diffusion size was found from the ratio (De/Z) and the Rich radius from (De_o/De) using the curves given in appendix B.

When high concentrations (i.e. $\geq$ Z 500 n/cm^3 and De $\geq$ 200 n/cm^3) were encountered, the photoelectric counter was used as a sensor. The technique is quite rapid. For low concentrations, the photographic Aitken counter was used, and multiple exposures were made to increase counting accuracy. This required an elapsed time of about 15 minutes to complete a series of observations, making it necessary to redetermine Z several times during the series to ensure that the total concentration is not varying through the measurement. While it would be desirable to have all measurements on the same aerosol sample, the relative homogeneity of maritime aerosol made this long time span seem less uncertain than the use of a bag or chamber for storage.

Aerosol measurements were made throughout the day, each and everyday of the voyage, beginning before, or just after sunrise, and continuing until after sunset. When the ship was under way, several measurements, including solar radiation measurements when the sun was visible, were made each hour. When the ship was on station, less frequent measurements were made. A majority of these observations were in uncontaminated maritime air; but additionally, measurements of aged continental aerosol were made in the English Channel, the North Sea, and off the Portugese Coast.

FIGURE I - CONCENTRATIONS OBSERVED
DEPARTING IBERIAN PENINSULA UNDER WEST
TO NORTHWEST WINDS - JUNE 1971



Results of Experiments

Many previous expeditions (Hogan et al.,1967) found continental aerosol overriding maritime aerosol several hundred miles offshore in the vicinity of the Bermuda and Azores high pressure systems. The aerosol concentrations off the east coast of North America have been found to hyperbolically approach maritime levels (Hogan 1970), and this implies that pure dilution of the continental air into the maritime air is the dominant force in reducing aerosol concentrations in these areas.

Measurements made upon leaving Lisbon Harbor and for 300 nautical miles offshore are plotted in Figure I, which shows a very rapid falloff to pure maritime air in less than 250 miles. This falloff is much more rapid than the hyperbolic rate found downwind of North America, and is undoubtedly due to the persistent north westerly winds which occurred during this experiment. Also, the "continental aerosol field" which usually surrounds the Azores Islands was not detected, presumably because the winds were northerly or light and variable while passing within 150 miles, to the north of the islands.

When all of the data is grouped as a function of distance from continental land at the time the observation was made, no simple relationship between distance from land and aerosol concentration is evident. When observations made less than 500 miles east of North America are analyzed, it is apparent that some residual continental aerosol was detected, but this continental effect was rapidly lost upon proceeding eastward.

The NOAA satellite nephanalyses were examined to investigate the possible influence of frontal systems on the aerosol concentrations observed. It was found that, when a storm system existed between the point of observation, and the North American Continent, relatively low (less than 300 $^{n}/cm^{3}$) concentrations of nuclei were found. Somewhat higher concentrations were found on the few occasions when no system was present.

When all of the concentrations observed between departing the main shipping lanes off the Portugese Coast, and re-entering them off the English Channel are grouped, the frequency distribution plotted as Figure II is obtained. When this frequency distribution is replotted as a cumulative distribution, the median concentration of 220 $^{n}/cm^{3}$ and slope of 1.5 are quite similar to those obtained in the Pacific, south of Hawaii by F. Guenther in 1968, but lower than values observed at 40^{o}N lat. during the last several years. When this median is compared to the averages of 650 $^{n}/cm^{3}$ obtained by Gunn (1964), and Landsberg's oft quoted oceanic average of 900 $^{n}/cm^{3}$, it seems very low; however, when compared to Landsberg's 1938 frequency analysis of the Carnegie data, which shows a median concentration of 255 $^{n}/cm^{3}$ it is remarkably close. This again

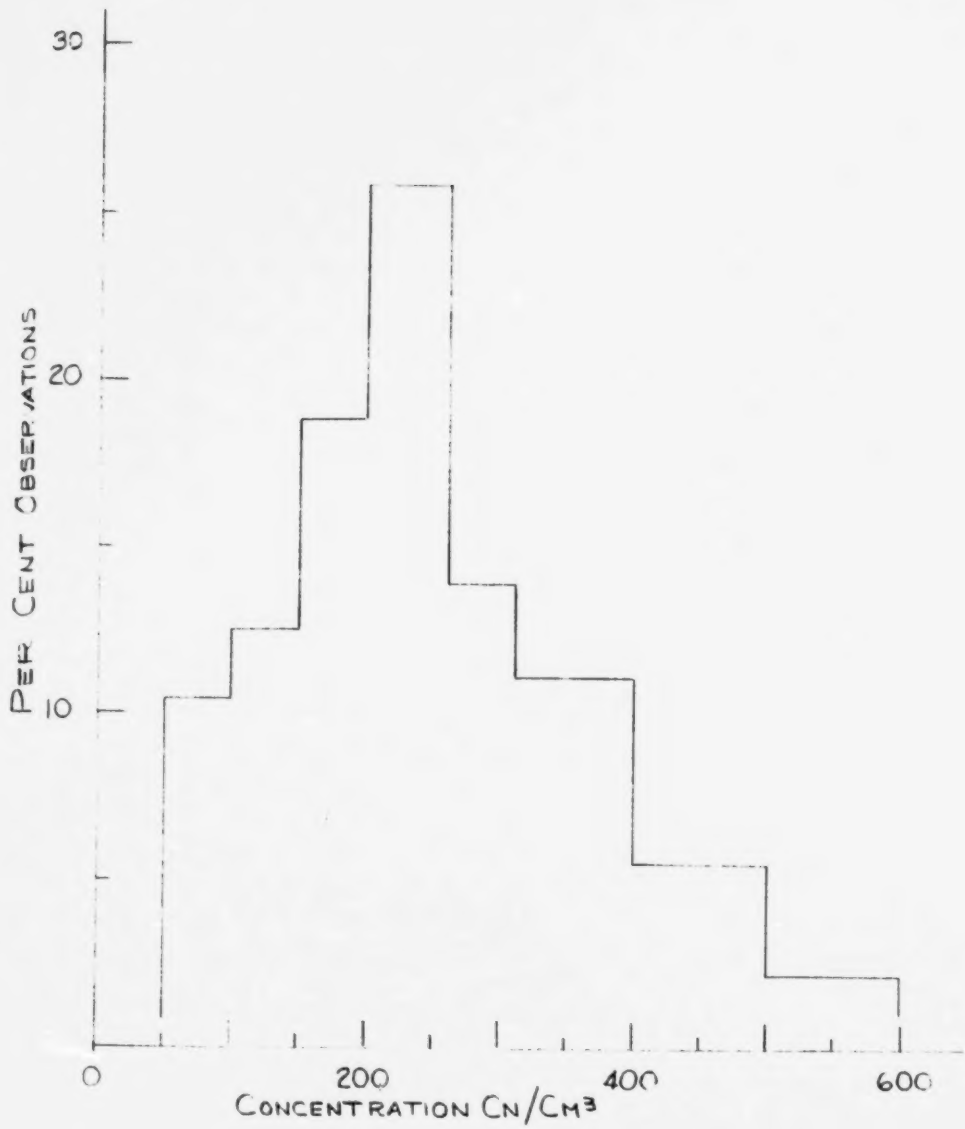


FIGURE II - FREQUENCY DISTRIBUTION
OF AITKEN NUCLEUS OBSERVATIONS MADE DURING
METEOR VOYAGE 23

shows the large difference in averages that can be obtained when only a few high readings are averaged with many low values.

It was observed, during Voyage 23, that extremely low (ie. less than 200 $^{n}/cm^{3}$) aerosol concentrations were found only at times of calm seas. The production of aerosol particles by breaking waves has been measured by Blanchard in Hawaiian surf, but mid oceanic production would seem to be less than that occurring in surf; also, the evaporation of saline droplets such as ejected from sea water, would appear to yield only a few large particles, rather than many small ones. An analysis was performed on the aerosol data, using wind data supplied by Dr. WURLITZER, meteorolgist of the Meteor. The data was grouped by wind classes, and the average concentration for each wind class was calculated. As few "high", (ie. > 400 $^{n}/cm^{3}$) values were observed during this voyage, little error is introduced by averaging, in this case . Average concentration is plotted against wind velocity in Figure III, which shows an increase in nucleus concentration with wind velocity, until the curve becomes constant at about 9 m/sec.. When the probability of finding a low concentration is calculated, this wind dependence is even more striking:

Wind Velocity M/sec	Percent of Observations $\leq 200/cm^3$	$\leq 175/cm^3$	$\leq 150/cm^3$	$\leq 125/cm^3$
0 - 4	91	63.5	54.5	36.4
4 - 6	52	30	22	13
6 - 8	40	29	24	7
8 - 10	33	19	19	5
10 - 12	50	12	12	0
12 - 14	0	0	0	0

As breaking waves begin to occur at 4.4 to 5.6 m/sec wind velocity, Röll, (1965) and the chances of finding concentrations less than 175 $^{n}/cm^{3}$ are small when this velocity is exceeded, it appears that breaking waves, through some mechanism, may be responsible in establishing a minimum aerosol concentration, over the seas. Many of these particles are undoubted sea salt crystal residue of bursting bubbles; however, the rapid oxidation (Allen and Day 1935) of dissolved gases released by these bubbles, or the conversion by some other chemical process, such as that which converts turpentine (Schaefer 1970) and other organics into particles may account for many more of these aerosol particles.

If photo chemical conversion of vapors to aerosol particles were present over the seas, one might expect to find a diurnal variation in aerosol concentrations similar to that found at remote

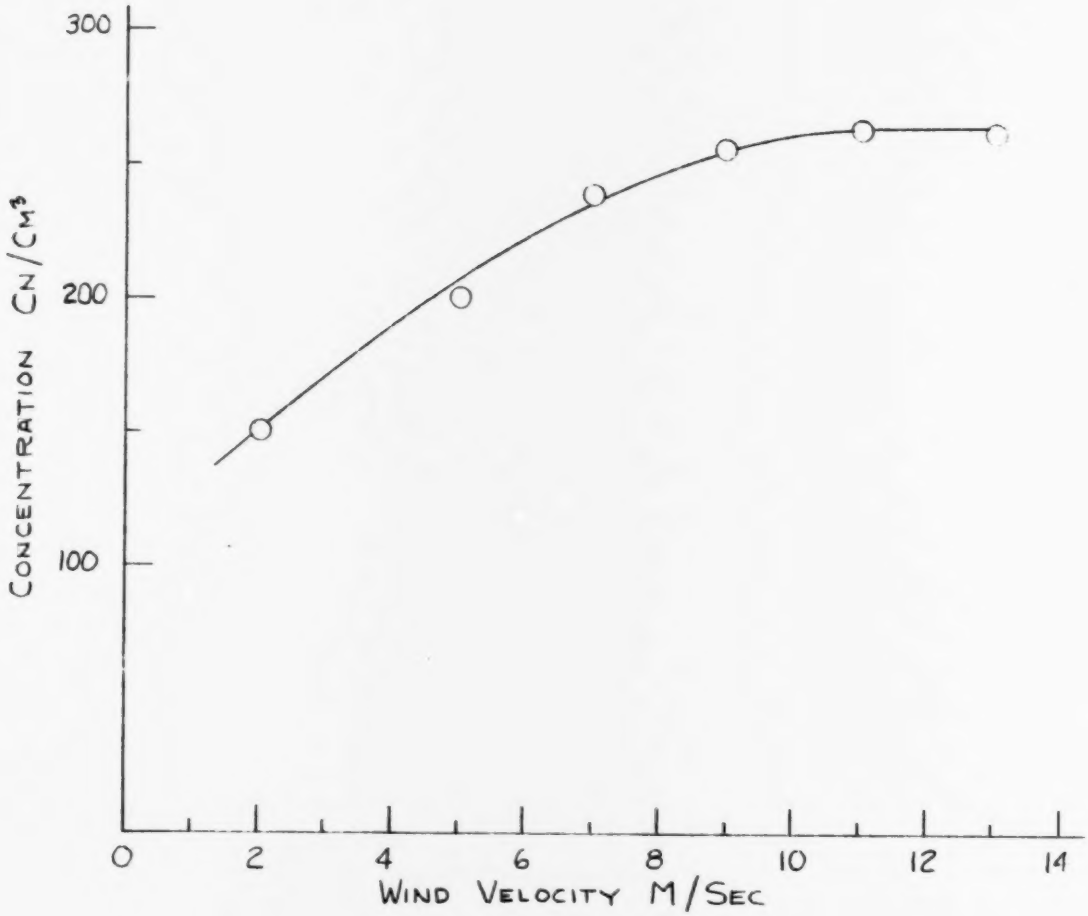


FIGURE III - AVERAGE VALUES OF
AITKEN CONCENTRATION VS WIND VELOCITY
AT TIME OF OBSERVATION

continental stations. When the observations made during Voyage 23 are grouped as a function of local sun time and averaged, as shown in Figure IV, no smooth diurnal tendency is found, in agreement with other investigators. When the aerosol concentrations are grouped in accordance with the meteorological conditions at the time of the observation, a significant pattern emerges:

Conditions at time of observation	Number observations	Average Concentration
Bright Sun	37	316 $^{n}/cm^{3}$
Weak Sun (Sufficient to cast shadows)	39	258 $^{n}/cm^{3}$
Sun not visible, no fog or precipitation	19	223 $^{n}/cm^{3}$
Sun not visible, rain	10	256 $^{n}/cm3$
Sun not visible, fog	9	166 $^{n}/cm3$
Sun not visible, precipitating fog or drizzle	22	146 $^{n}/cm^{3}$
All observations	136	245 $^{n}/cm^{3}$

The average values observed in bright sun are 1.29 times the average obtained for all observations in maritime air, and 1.22 times the average of observations made in weak sun. This indicates that solar conversion of vapors to aerosol particles may well be present over the sea, and that the lack of a smooth diurnal variation over the sea may be due to the banded nature of clouds and fog patches, rather than to the lack of solar influence.

This tabulation also shows the effectiveness of several processes which remove aerosols from the atmosphere near the surface. Rain (meaning moderately large falling drops) is relatively ineffective in reducing the number concentration of small aerosol particles in the air through which it falls. Fog, and especially precipitating fog or drizzle, consisting of extremely small droplets extending to the surface of the sea, is a strong mechanism in reducing the number concentration of aerosols. This is probably due to the falling drops removing the active cloud condensation nuclei, the large surface area of the slow moving drops presenting a good target for diffusional collection of particles, and some electrostatic collection of charged particles.

Surprisingly, the smallest equivalent sizes of aerosol particles were found during these heavy fogs and drizzles. From this it can be inferred that fog is active in removing the larger, more meteorologically active aerosol particles.

When the sizes obtained for the aerosol throughout the voyage are analyzed, only two general patterns emerge; the maritime aerosol is larger than the continental aerosol found in uncontaminated areas and the aerosol became progressively smaller in the vicinity of the continent. The equivalent diffusion radius was greater than 1.5×10^{-6} cm in 75% of observations and the Rich equivalent radius was greater than 3×10^{-6} cm 63% of the time, in mid ocean

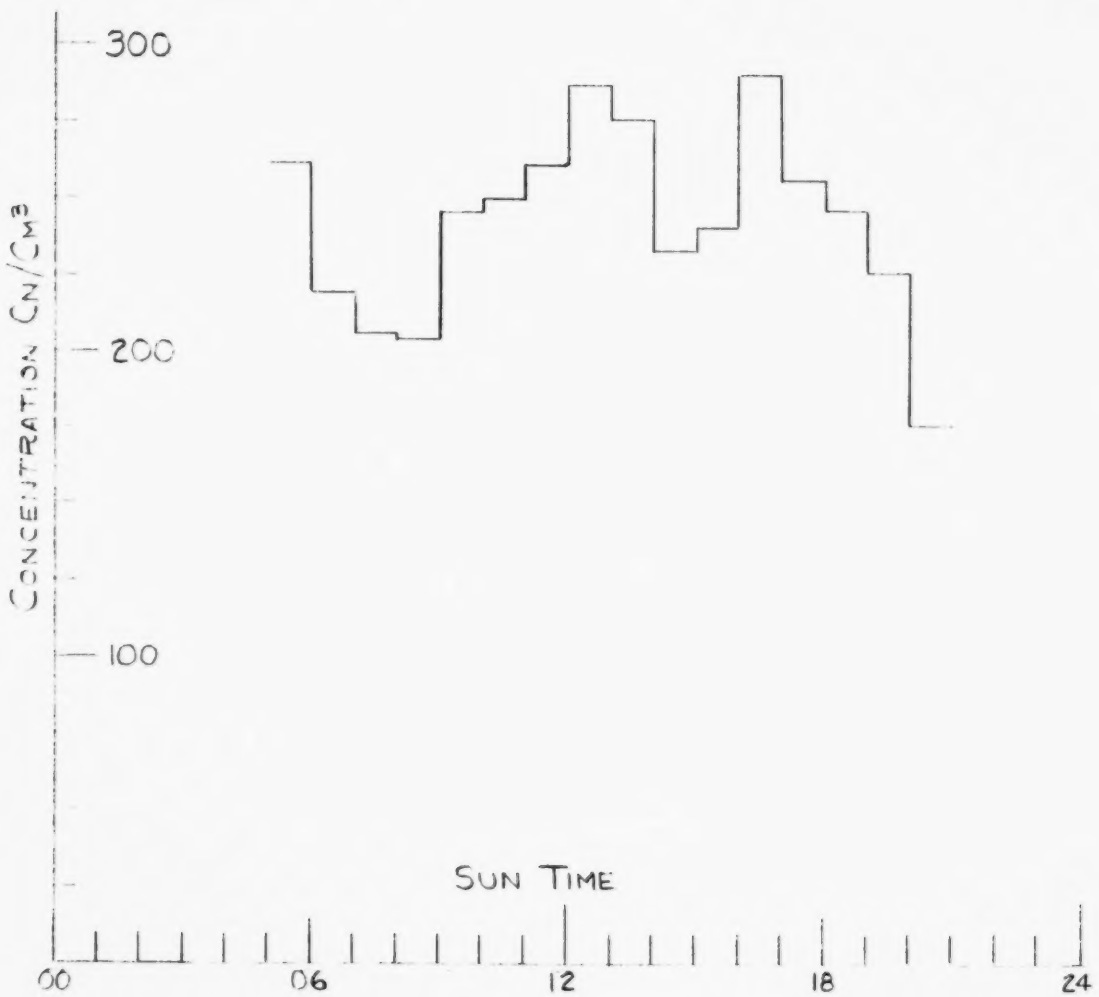


FIGURE IV - AVERAGE AITKEN CONCENTRATION
vs TIME OF OBSERVATION, METEOR VOYAGE 23,
11 JUN - 24 JUN 1971

This can be interpreted as an absence of extremely small particles, which would be expected in an area of low aerosol production and is a characteristic of an "aged" aerosol. The aerosols encountered off the Portugese Coast, and in the North Sea generally had diffusion radii less than 1.5×10^{-6}cm, and Rich radii less than 3.0×10^{-6} cm showing less evidence of "aging". The aerosols measured in Lisbon Outer Harbor were very small, with diffusion sizes of less than 1.0×10^{-6} cm, even though the concentration was very low (less than 1500 $^{n}/cm^{3}$) for a continental area; evidently the strong (10 ^{m}/sec) offshore (249^{o}) winds were rapidly diluting fresh aerosols to this level, with little or no coagulation aging.

The sizes obtained during the voyage must be considered preliminary, and the ratios of aerosol concentation transmitted in the various modes approach the resolution of the counting instrument; in many cases the diffusion loss could not be resolved. As the aerosol size over the seas is a major unknown these size measurements should be repeated with a slightly modified photographic Aitken Counter of greater resolution.

Conclusions

A region of low (i.e., < 300 $^{n}/cm^{3}$) Aitken nucleus concentrations exists over the North Atlantic, north of 40^{o}N latitude. This low concentration region extends to the mouth of the English Channel under favorable meteorological conditions.

No smooth diurnal variation of Aitken nucleus concentrations existed in the region studied during the time of the voyage. Aerosol concentrations were found to be highest at times of bright sun; as bands of cloudiness exist in this area, such intermittent sunshine may mask a solar diurnal for such a short period of time.

The lowest aerosol concentrations were found when fog, precipitating fog, and drizzle were observed concurrent with aerosol measurement, and sustained concentrations of greater than 300 $^{n}/cm^{3}$ were found only when no frontal system was present between the North American continent and the ships' position. This indicates that the physical processes within the cloud are active in reducing aerosol concentrations to the levels usually associated with maritime air.

Extremely low concentrations (i.e., 175 $^{n}/cm^{3}$) were found only in the absence of crested waves, but the average number concentration of particles ceased increasing as a function of wind when a wind velocity of 9 ^{m}/sec was reached. A maritime background of 150 to 250 $^{n}/cm^{3}$ can apparently be mainted by wave action in the absence of other sources.

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APPENDIX D

INSTRUCTIONS FOR COOPERATING OBSERVERS

APPENDIX D

GENERAL INSTRUCTIONS FOR

AEROSOL OBSERVATIONS AT SEA

1. Observations will be made in conjunction with NOAA Weather Bureau 6 hourly observation.

2. Observations will be made from upwind wing of Flying Bridge, unless the observer believes that less contaminated air can be obtained on some other point on the superstructure.

3. Each observation will consist of four (4) individual readings of the instrument.

4. Observations will be recorded on a carbon copy of the NOAA observation sheet.

5. Ventilating funnels, certain cargos containing creosote or turpentine, and cigarettes, generate large quantities of aerosol particles, sometimes too small to be seen as a visible cloud of smoke. These sources are to be avoided when making observations; if some other types of odiferous cargos appear to produce measurable aerosols the information will be of interest to the investigators.

6. Please note interesting meteorological phenomena (nearby lightning, St. Elmo's fire, waterspouts, Northern Lights, clouds) on the report sheet as convenient.

7. Expected ranges of Aerosol Concentration

In Port	-	30,000 n/cm^3 or more
Near Shore	-	3,000 n/cm^3 - 30,000 n/cm^3
Well off shore	-	1,000 n/cm^3 - 3,000 n/cm^3
Mid Ocean	-	less than 1,000 n/cm^3

These numbers are offered as a guide to sampling site location on the ship's superstructure, the range in nature is sometimes much greater. A good indication of freedom from contamination is $\pm$ 1 scale division repeatability of observations.

8. Records will be mailed upon arrival at a U. S. Port to:

A. W. Hogan
Atmospheric Sciences Research Center
130 Saratoga Road
Scotia, New York 12302

9. Call A. W. Hogan, at 518/377-6477, collect from any U. S. Port. If questions arise, Dr. Degani or Cadet Assistants can be reached at TY-2-3000.

INSTRUCTIONS FOR NORTH ATLANTIC OBSERVATIONS

It has been observed that continental pollution, in the form of combustion aerosols (called "Condensation Nuclei", "Aitken Nuclei" or just "Particles") extends 1000 miles offshore in the Atlantic between 20^{o}N and 40^{o}N. It appears that this is not the case above 40^{o}N, even though prevailing currents would lead one to believe that the path of combustion particles from the populous east coast would be along the coast and out to sea in the area 40^{o}N to 60^{o}N by 60^{o}W to 40^{o}W.

The frequent precipitation which occurs above 40^{o}N, as opposed to the scarcity south of 40^{o}N is apparently responsible for the removal of these particles, although other mechanisms may also be active. We are studying the aerosol concentrations over the North Atlantic in an attempt to understand these processes which nature uses to clean the air. In order to study these processes, we need many aerosol observations made concurrently with meteorological observations unders as many weather conditions as is possible, along the entire Atlantic Trade Route.

Observations are to be made as part of the 6 hour weather Obs, and will consist of 4 readings with the Gardner instrument. The values obtained will be entered in the remarks column of a carbon copy of the weather bureau data sheet.

All combustion processes, (including cigarettes and ship's funnels) and some chemical vapors (including paint thinner and gasoline) produce particles. It is therefore important to find a spot on the ship's superstructure free from local contamination to make observations. Usually the upwind wing of the Flying Bridge is a good location, but, depending on wind direction and velocity, and ship's speed, this too can become contaminated. The following rules of thumb can be used to detect local contamination:

1) Local contamination is not well mixed, and therefore gives unsteady, erratic readings.

2) Local contamination always raises values. The expected values are approximately:

   a) N.Y. Harbor, L.I. Sound, Coastal waters near cities--50,000 to 250,000 $^{n}/cm^{3}$ (50-80 scale Div)

   b) Offshore--1,000 to 50,000 (10-50 scale div) depending on distance offshore.

   c) Mid Ocean Maritime--<1000 $^{n}/cm^{3}$ (0-10 scale div)

FIELD TECHNIQUES FOR IMPROVED RELIABILITY

IN AITKEN NUCLEI MEASUREMENT

INTRODUCTION:

The small particle detector, patented by Rich of General Electric and currently manufactured by Gardner Associates is a rugged field instrument, of high reliability and good repeatability. It does, however, require that a consistent technique be applied in its operation to ensure this high standard of performance. The suggestions herein result from experience gained from several experiments in varying environments, and from a short series of chamber experiments with a 5000 litre test chamber.

GENERAL CONSIDERATION FOR CARE OF THE PARTICLE COUNTER.

Although the Gardner counter is very rugged and can withstand the rigors of transportation very well, it is sensitive to seemingly minor mistreatment during routine maintenance and storage. The photocell undergoes an apparent change in sensitivity if it received no light for a long period, or if it is exposed to bright lights. This is only a short term effect, (order of days) but it can be avoided by occasional periods of exercise of the instrument during storage, and by using the following technique when moistening the fog tube: dim the lights, or go into a shaded area, (as you would when loading a camera), when removing the fog tube for moistening. Cover the tube entrance with a card or book when the tube is removed, or, if the entire plate is removed for maintenance, cover the photocell itself.

Always inspect the blotting paper lining for tears or defects when moistening. These can cause shadows which will effect accuracy. Also, when moistening, be sure that no liquid water remains on the surface of the blotter when the tube is reassembled. This can be ensured by moistening the blotter thoroughly, and allowing the tube to stand upright for 3 - 5 minutes, then shaking the tube as if it were a clinical thermometer, and standing it upright again for 3 - 5 minutes. If no liquid droplets appear at the bottom of the tube, it is ready for assembly; if not, repeat the shaking and standing procedure until no drops appear.

Battery maintenance is minor. The high voltage supply should be replaced annually on a routine basis. The $1\frac{1}{2}$ V battery supply should be replaced whenever frequent adjustment of the "coarse" potentiometer is necessary to achieve "zero".

TECHNIQUE OF MAKING A MEASUREMENT.

The following technique of making a measurement with the Gardner Associates Counter was developed in the field, and has been tested against a stable aerosol supply (the 5000 litre chamber). With practice, it is possible to make readings at a rate of one reading per minute with this technique. It is most important that the technique be followed in the same order and at the same speed each time, so that diffusion losses inside the instrument will be consistent.

TECHNIQUE FOR DETERMINING AITKEN COUNT WITH PORTABLE COUNTER.

Preliminary

1. Roll adjustment potentiometers full rotation three (3) times. (if counter has not recently been used.)

2. Switch on counter and draw air through the instrument with the pump, using about 15 pump strokes.

3. Adjust the "FINE" potentiometer to the point of maximum control (i.e., about one-half to two-thirds of the resistance in the circuit), then adjust the pointer with the "COARSE" potentiometer.

4. Operate the hand pump while watching the pointer. Re-adjust the "fine" and "coarse" adjustments if necessary to achieve "zero".

5. Perform a complete sampling operation as shown in the instruction manual. Note the reading, open the valves and purge with the hand pump. The zero should repeat within one scale division. If, after several readings, zero fails to repeat, the sequence of preliminary operations should be repeated.

GENERAL PROCEDURE FOR MAKING A MEASUREMENT AFTER WARM-UP.

1. Draw 7 to 10 strokes of sample through with the hand pump. Then repeat zero technique (above). Ensure that the instrument is completely purged; if it requires more than 10 strokes, contamination may be present.

2. When sealing the valves in sequence, do so gently; snapping the valves can change the contact resistance of the potentimeter causing a zero shift.

3. Open valve V3, to create the expansion, with a firm, steady push. (Experiments with an oscilloscope on the photocell output show that the shape of the extinction-time curve can be influenced by the way the valve is opened.

4. Read the highest rise of the pointer, then reopen valves and purge. Doubt any reading which is not followed by a consistent zero.

SPECIAL INSTRUCTIONS FOR MAKING OBSERVATIONS IN THE FIELD.

1. Extreme cold. It is best to keep the instrument in a warm location, and connect to the free atmosphere with 4-6 feet of ¼" tygon tubing. In an automobile, it is necessary to wait 15 minutes for the contamination due to the engine to subside, then extend the tubing about 2 feet out of the window, upwind. The instrument has been employed below $0^{\circ}F$, but performance is impared by the operators' inability to accurately zero the instrument under these conditions. Rain has no effect on measurement, if liquid water is not permitted to enter the instrument.

2. Fog. Extreme care must be taken when experimenting in fog, as the number of fog droplets occuring in nature, will be of the same magnitude as the number formed in the fog tube. If the instrument is slightly warmer than the air, there is less chance for condensation to occur in the photocell.

3. Aboard ship. The instrument itself does not seem to be affected by the ship's oscillation, but the indicator pointer sometimes oscillates with the roll of the ship. This can be minimized by orienting the indicator at right angles to the plane of severest motion.

4. In general, it is best to keep the instrument in shelter, and warmer than the ambient air, until shortly before the observation is made.

APPENDIX E

DATA USED TO CONSTRUCT FIGURES AND TABLES

K•E 4 X 4 TO THE INCH 46 0253
7 X 10 INCHES MADE IN U.S.A.
KEUFFEL & ESSER CO.

NUMBER OF OBSERVATIONS

WEST LONGITUDE

80 70 60 50 40 30 20 10 0

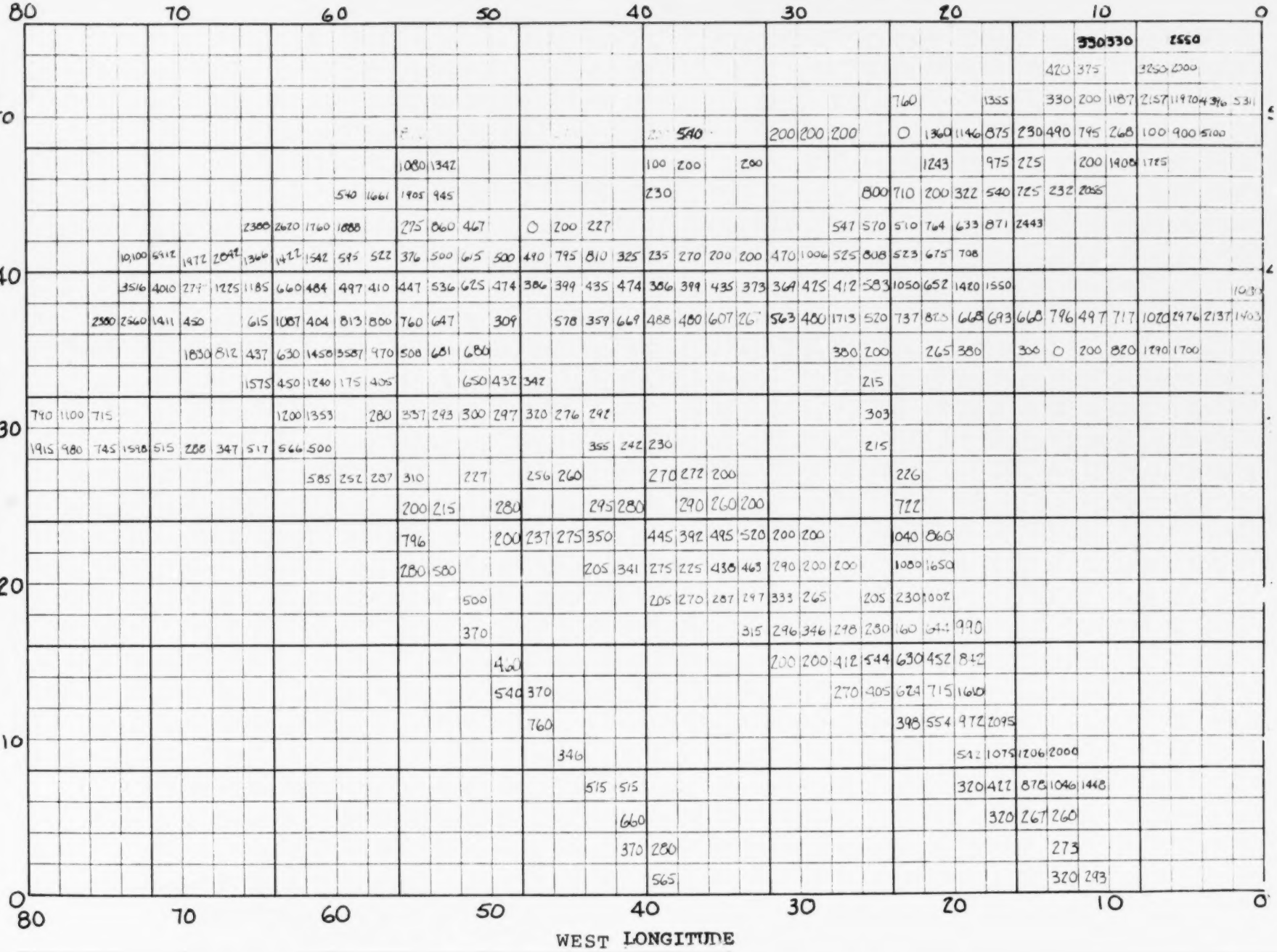
NORTH LATITUDE

50 40 30 20 10 0

93

K•E 4 X 4 TO THE INCH 46 0253
7 X 10 INCHES MADE IN U.S.A.
KEUFFEL & ESSER CO.

AVERAGE NUCLEUS CONCENTRATION OBSERVED



K•E 4 X 4 TO THE INCH 46 0253
7 X 10 INCHES MADE IN U.S.A.
KEUFFEL & ESSER CO.

NUMBER OF OBSERVATIONS USED TO CONSTRUCT TABLE 2 - SECTION VI

		<100	<200	<300	<400	<500	<600	<800	<1000	<1200
6 JUN	14 JUN	0	1	16	15	10	10	12	3	0
4 JUL	10 JUL	0	0	12	10	15	10	12	2	2
19 JUL	26 JUL	0	5	31	10	10	5	8	3	0
16 AUG	22 AUG	0	0	10	16	24	3	6	1	0
SUMMER	1970	0	6	69	51	59	28	38	9	2